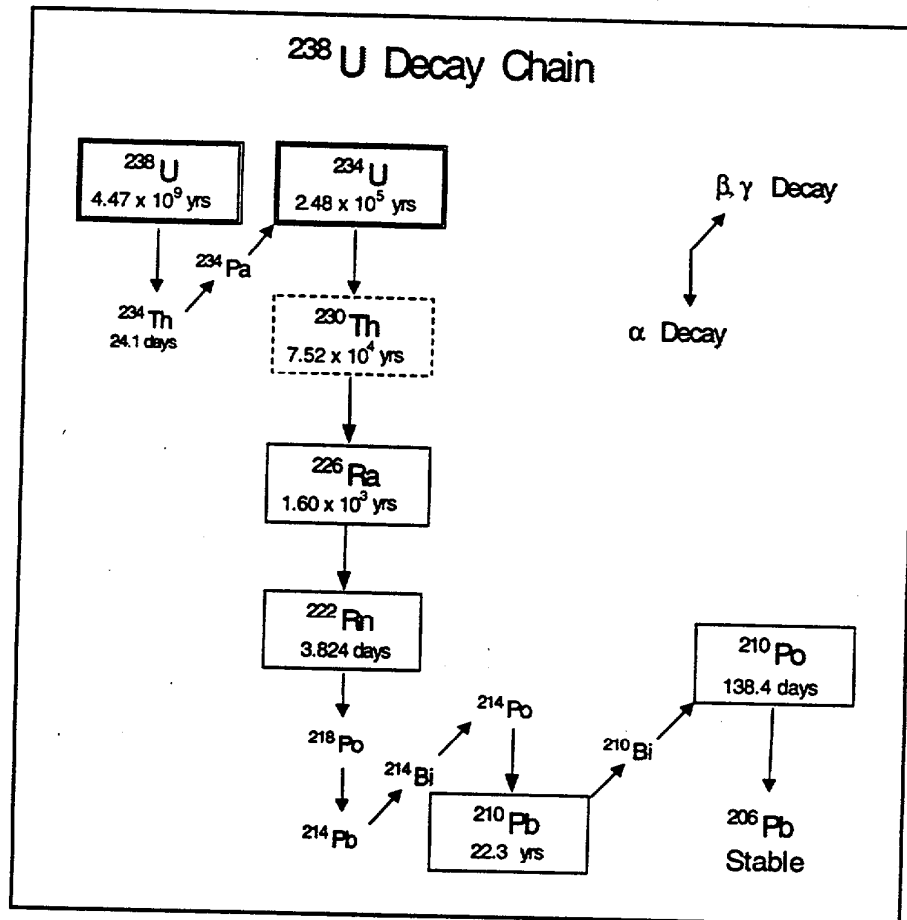


# NORM and TENORM

(Naturally Occurring and Technologically Enhanced  
Naturally Occurring Radioactive Material)

## Producers, Users, and Proposed Regulations



### PEP Course 1.A Notes

Health Physics Society 1999 (32<sup>nd</sup>) Midyear Meeting  
Albuquerque, New Mexico  
24 January 1999

Notes Prepared by:

Phillip Egidi and Carter Hull

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Notes Prepared by:

**Phillip Egidi**

Technical Coordinator, Environmental Technology Section  
Health Sciences Research Division, Oak Ridge National Laboratory  
P.O. Box 2567, Grand Junction, Colorado 81502  
(970) 248-6189  
7pe@ornl.gov, pe@normis.com  
www.normis.com

and

**Carter Hull, Ph.D., Ph.D., J.D., M.S., R.G., P.G.**

Radiochemistry Program Director  
Oxford Instruments, Inc., Nuclear Measurements Group  
601 Oak Ridge Turnpike, Oak Ridge, Tennessee 37830  
(423) 482-4041 x226  
chull@oxfordnuclear.com  
carterhull@aol.com  
www.oxfordnuclear.com

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## INTRODUCTION

Naturally Occurring Radioactive Material (NORM) is everywhere. We are exposed to it every day. However this is not unusual; it is, as the terminology expresses, natural. Actually, NORM is one of the primary reasons that Earth remains habitable. NORM is the source of the Earth's heat flux, drives the movements of the planet's tectonic plates, and so plays a major rôle in the evolution of life on Earth. NORM is an integral part of the planet, our bodies, the food we eat, air we breathe, the places where we live and work, and within products we use. We are also bathed in a sea of natural radiation coming from the sun and deep space. Terrestrial life has evolved with this natural radiation and radioactivity. Conversely, processing of some natural resources concentrates naturally occurring radionuclides to a degree that they may pose risks to humans and the environment. These and other activities, such as flying at high altitudes, increase exposures to radioisotopes beyond limits that occurred naturally.

This session focuses on Technologically Enhanced (TE) NORM. All degrees of TENORM exist, but our concerns are typically with components of large-volume, low specific radioactivity industrial waste streams. These include minerals mining, transport, beneficiation and chemical processing of ores, production of phosphate fertilizers, extraction and purification of trace elements, manufacture of some metals, water treatment and purification, use of TENORM-bearing by-products, as well as oil and gas and other energy production. The majority of radionuclides in TENORM are found in the uranium and thorium decay chains. Radium ( $^{226}\text{Ra}$ ) and its decay products (*e.g.*, radon –  $^{222}\text{Rn}$ ) are quite often used to characterize the redistribution of TENORM that results from human activities. A synopsis of these large volume streams of TENORM will be presented, as shall a brief review other natural radionuclides, *e.g.* potassium and rubidium isotopes, and other such minor sources that primarily contribute to background doses.

TENORM is found in various concentrations in a variety of forms (physical and chemical matrices) such as scrap metal, sludges, slags, fluids, scales in storage tanks and piping, chemical residues, processing fluids, surface and groundwaters, and mine tailings. TENORM has also been found in industries that were not thought to be subject to radioactive contamination. The levels

of and sources of radioactivity in TENORM vary significantly in various forms. Although TENORM is prevalent in industrialized societies, it is emphasized that few industrial processes actually concentrate NORM. Neither the Atomic Energy Act nor any other U.S. Federal regulations address TENORM. Control and regulation of TENORM is inconsistent from industry-to-industry and between states, provinces, and nations.

So when is TENORM a problem? Where is it a problem? This depends on who is presenting and organizing the data and discussion, when it is presented, and where. One is likely to receive somewhat differing views on exactly the same topic from, for instance, the phosphate industry *versus* that of state/provincial or federal regulators. The authors do not wish to contribute to conflicting views on TENORM regulatory issues; our main objective is to present unbiased facts relating to NORM and TENORM. These session notes are organized to begin with a brief review of background radioactivity followed by a discussion of the geological framework, mobility, and variability of these radionuclides. A review of some of the industrial sectors affected by TENORM is then given. An overview of proposed guidelines and regulatory aspects of TENORM concludes these notes.

## BACKGROUND RADIOACTIVITY

*Ceiling Tiber Hat  
U - Radon  
Helite*

There are two generic definitions for background radioactivity:

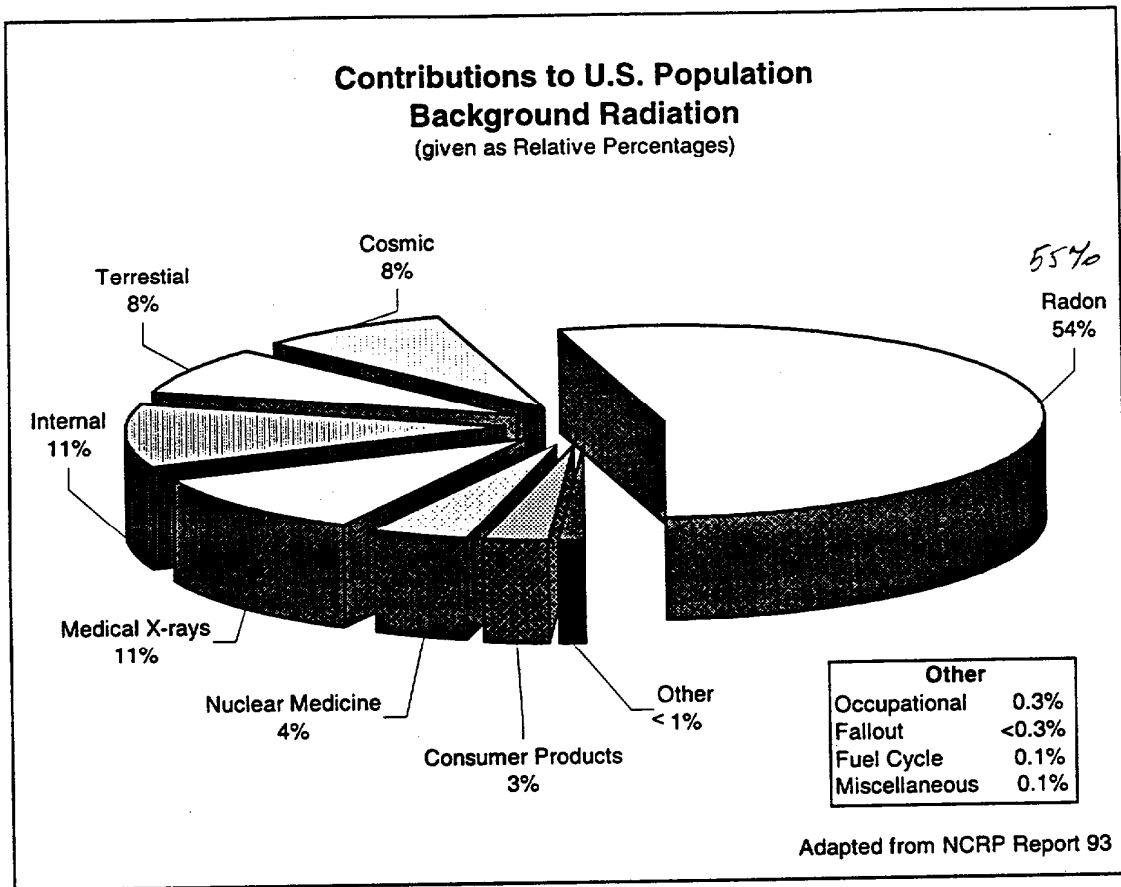
- 1) Radioactivity that is the result of naturally occurring concentrations of radionuclides that represent ambient conditions present in the environment that are *in no way influenced by human activity*, or
- 2) Concentrations of radionuclides from *anthropogenic sources originating from non-site sources* (Gesell and Prichard, 1975). An example is global fallout.

## Components of Background Radioactivity

The following discussion is drawn largely from NCRP 94. Four major components constitute "background sources" of radiation:

- Human Produced
- Terrestrial
- Cosmogenic
- Cosmic

Most TENORM is associated with terrestrial sources, but the other types may interfere with measuring levels of TENORM. Fig. 1 shows the background sources to the U.S. population.



**Figure 1. Sources of Background Radiation to the Public**

### **Human Produced (Anthropogenic) Sources of Radioactivity**

Anthropogenic sources of radiation are often considered as a component of the background. Why is this; it is not logical or intuitive? This likely occurred since these types of human produced sources are ubiquitous throughout the environment as opposed to being locally distributed. Activities that have contributed to the dispersion of radionuclides in the environment include:



- Nuclear weapons tests and use
- Nuclear accidents (Chernobyl) and incidents
- Nuclear reactors (for this discussion,  $^{14}\text{C}$  and Tritium)

Most anthropogenic radionuclides are short-lived, but some have intermediate half-lives of a few years and worthy of note. These intermediate half-lived nuclides are listed in **Table 1**.

**Table 1. Examples of Half-Lives for Some Anthropogenic Radionuclides**

Radionuclides of Interest	Half-Life ( $t_{1/2}$ )
$^{137}\text{Cs}$	30.2 y
$^{90}\text{Sr}$	28.1 y
$^{85}\text{Kr}$	10.73 y

Also, the global inventory of  $^{14}\text{C}$  and  $^3\text{H}$  has been increased from human activities, and it is sometimes necessary to measure these globally distributed radionuclides separately and to distinguish them from locally produced sources. In addition Pu isotopes were released in fallout.

The variability of anthropogenic sources of radiation and radioactivity relates directly to the population distribution and level of technology found in different areas around the world. Deposition in an area depends upon wind and precipitation patterns (NRC 1994).

### Cosmic Radiation

This type of background refers to both the *primary* energetic particles of extraterrestrial origin that strike the earth's atmosphere and to the *secondary* particles generated by their interaction with the atmosphere.

**Primary radiation** itself consists of two components, designated as *galactic* or *solar* depending on origin.

Primary particles are attenuated in upper atmosphere. Reactions take place and generate secondary particles. The cosmic radiation field at ground altitude (0 to 3 km) consists almost entirely of secondary particles whose origins are almost exclusively galactic.

Annual external dose rates from cosmic rays depend slightly on latitude and strongly on altitude (Table 2). The latitude effect is due to the charged-particle nature of the primary cosmic rays. When they come near the earth, its magnetic field tends to deflect the rays away from the equator and toward the poles (Gollnick 1988).

**Table 2. Altitude Dependence of Cosmic Ray Dose**  
(Dose equivalent; does not include the neutron component).

Altitude, m (ft)	Dose Rate, $\mu\text{Sv yr}^{-1}$ (mrem $\text{yr}^{-1}$ )	Example
Sea level	~270 (31)	Los Angeles
1,525 (5,000)	~478 (55)	Denver
3,050 (10,000)	~1,190 (137)	Leadville, Colo.
9,140 (30,000)	~16,530 (1900)	Normal jetliner
15,240 (50,000)	~76,125 (8750)	Concorde
24,340 (80,000)	~106,140 (12,200)	Spy plane

Adapted from Gollnick 1988.

## Cosmogenic Radiation

Cosmogenic radionuclides arise from the collision of highly energetic cosmic ray particles with stable elements in the atmosphere and in the ground. The entire geosphere, the atmosphere, and all parts of the earth that directly exchange material with the atmosphere contain cosmogenic radionuclides. The major production of cosmogenic radionuclides results from the interaction of cosmic rays with atmospheric gases.

The outermost layer of the Earth's crust is another area where reactions with cosmic rays occur. However, the rate at which they occur is several times smaller than the atmospheric component because most of the cosmic rays are attenuated in the atmosphere. The result is that the contribution to background dose is minimal.

The most important cosmogenically produced radionuclide is  $^{14}\text{C}$ . However, many others, such as  $^3\text{H}$ ,  $^{22}\text{Na}$ , and  $^7\text{Be}$ , occur. Carbon-14 produced in the atmosphere is quickly oxidized to  $\text{CO}_2$ . The equilibrium concentrations of  $^{14}\text{C}$  in the atmosphere are controlled primarily by the exchange of  $\text{CO}_2$  between the atmosphere and the ocean. The oceans are the major sinks for removal of  $^{14}\text{C}$  from the atmosphere.

Most of the other cosmogenically produced radionuclides in the atmosphere are oxidized and become attached to aerosol particles. These particles, or geoaerosols, act as condensation nuclei for the formation of cloud droplets and eventually coagulate to form precipitation. About 10 to 20% of cosmogenically produced radionuclides are removed from the atmosphere by dry deposition on the earth's surface.

Concentrations of cosmogenic radionuclides vary in the atmosphere with time and location. Variations are day-to-day, seasonal, longitudinal, and sunspot-cycle related. The concentrations of some cosmogenic radionuclides, such as  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{22}\text{Na}$ , and  $^{37}\text{Ar}$ , have increased during nuclear tests. Reactors also generate  $^{14}\text{C}$  that eventually will be distributed in the atmosphere, but is estimated to be two orders of magnitude lower than the natural concentration.

The total effective dose equivalent rate to the body produced by the primary cosmogenic radionuclides is just over 10  $\mu\text{Sv}$  (1 mrem  $\text{yr}^{-1}$ ), with essentially the entire dose arising from  $^{14}\text{C}$ .

## Terrestrial Radiation

The final component of background comes from radionuclides found on Earth. Several dozen naturally occurring radionuclides have half-lives of at least the same order of magnitude as the estimated age of the earth ( $4.5 \times 10^9$  y), and are assumed to represent a primordial inventory. These primordial radionuclides are also what we are most concerned with in the TENORM issue.

The primordials are usually divided into two groups:

- Those that occur singly (non-series) and decay directly to a stable nuclide
- Those radionuclides that occur in decay chains (series) and decay to a stable isotope of lead through a sequence of radionuclides of wide-ranging half-lives.

## Non-Decay Series Radionuclides

Two primary non-decay series radionuclides contribute to background dose, Potassium-40 ( $^{40}\text{K}$ ) and Rubidium-87 ( $^{87}\text{Rb}$ ). Potassium-40 is a beta (87.3%) and gamma (10.67%) emitter and contributes to both internal and external doses. It exists as a constant fraction of stable potassium (0.0117%). Potassium-40 behaves chemically as does stable potassium. It is ubiquitous at the Earth's surface and is concentrated in felsic igneous rocks and their weathering products. Potassium-40 is also concentrated in flowering plant buds, in cereal grains, and mesostems of many plants. Potassium-40 is also present in most organisms; for instance human sweat contains  $^{40}\text{K}$ . This is one reason why gamma spectrometrists take care not to touch detectors or sample containers – an elevated background for the  $^{40}\text{K}$  photopeak may result. The  $^{40}\text{K}$  contribution to

external dose is variable, depending on its concentration in rocks and soil. Average concentration is about  $0.6 \text{ Bq g}^{-1}$  ( $17 \text{ pCi g}^{-1}$ ) in crustal rock. Potassium-40 is found in TENORM, particularly building materials (bricks, cinder blocks). It may be necessary to determine background fractions separately from total concentrations. Potassium is metabolically regulated by the body and is not controlled by intake.

Rubidium-87 is a pure beta emitter and is found in crustal rock in concentrations of about  $0.07 \text{ Bq g}^{-1}$  ( $2 \text{ pCi g}^{-1}$ ). It is not an external hazard and is rarely considered in dose calculations. However,  $^{87}\text{Rb}$  is quite important in the generation of heat in the mantle and crust of the Earth.

The remainder of the non-series radionuclides has combinations of half-lives, isotopic abundance, and elemental abundance such that they have negligibly small specific activities and are not significant in background calculations.

### Decay Series Radionuclides

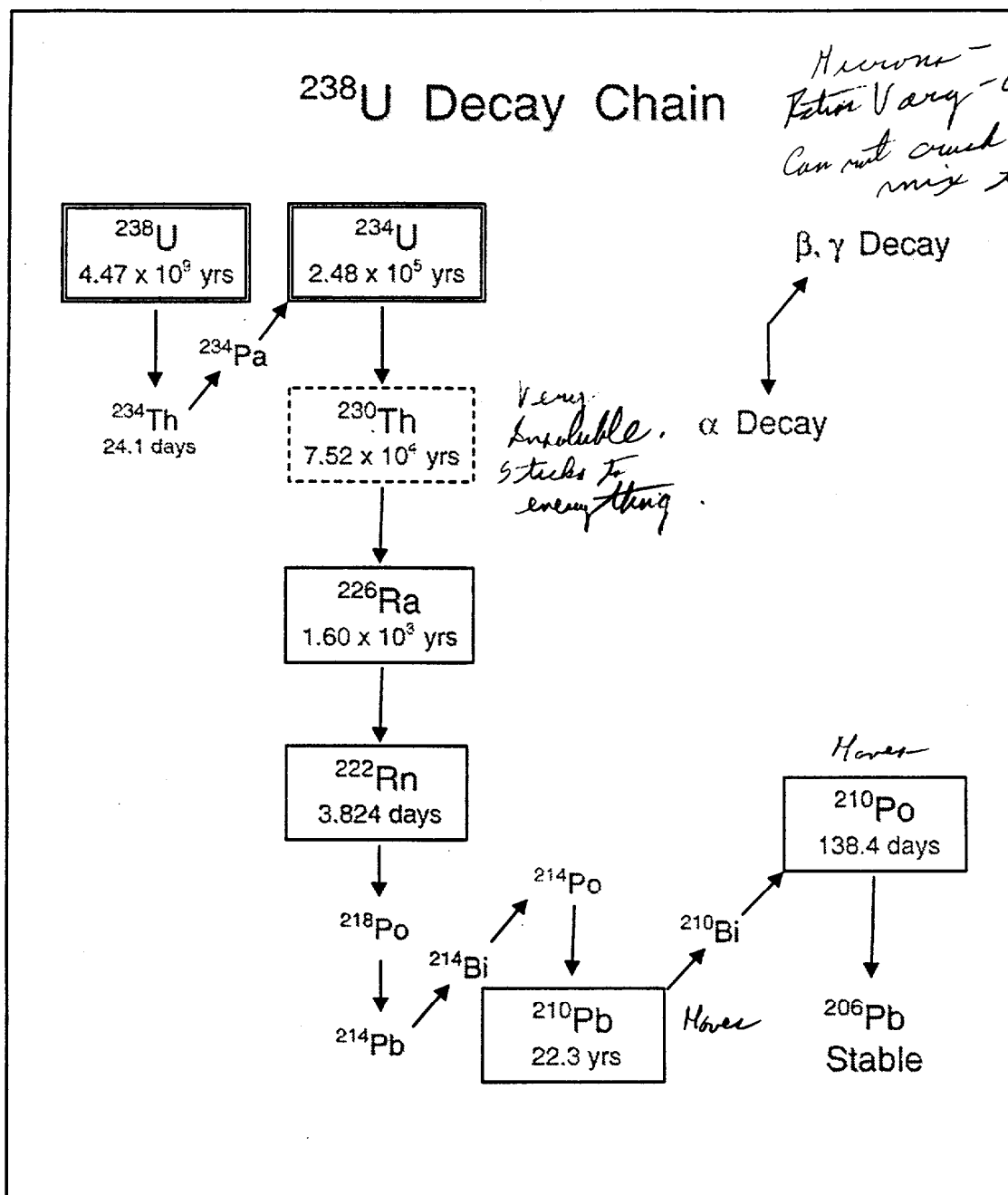
There are three naturally occurring decay series, headed by the radionuclides  $^{238}\text{U}$ ,  $^{235}\text{U}$ , and  $^{232}\text{Th}$ . These series are commonly called the uranium series, the actinium series, and the thorium series respectively. **Table 3A** lists components of the uranium and thorium series. **Table 3B** lists a few of the non-series radionuclides. Generally, the actinium series does not play a significant rôle in industrial TENORM due to its very low presence (one-sixth of  $^{238}\text{U}$ ) in the natural environment.

If not subjected to chemical or physical separation, each of these series attains a state of secular radioactive equilibrium. Technological enhancement of NORM as well as natural physical and chemical reactions often interfere with this balance. Crustal concentrations of the heads of the three series are extremely small (parts *per* million); the short-lived decay progeny are present in such exceedingly minute concentrations that their behavior does not always follow chemical (mass action) controls. There will be further discussion about this later.

Table 3A. Principal Natural Radionuclide Decay Series

Nuclide	Symbol	Half-Life ( $t_{1/2}$ )	Major Radiations
<b><u>Uranium-238</u></b>	$(^{238}\text{U})$	4.47 billion years	alpha, x-rays
Thorium-234	$(^{234}\text{Th})$	24.1 days	beta, gamma, x-rays
Protactinium-234m	$(^{234\text{m}}\text{Pa})$	1.17 minutes	beta, gamma
Uranium-234	$(^{234}\text{U})$	248,000 years	alpha, x-rays
Thorium-230	$(^{230}\text{Th})$	77,000 years	alpha, x-rays
Radium-226	$(^{226}\text{Ra})$	1600 years	alpha, gamma
Radon-222	$(^{222}\text{Rn})$	3.83 days	alpha
Polonium-218	$(^{218}\text{Po})$	3.05 minutes	alpha
Lead-214	$(^{214}\text{Pb})$	26.8 minutes	beta, gamma, x-rays
Bismuth-214	$(^{214}\text{Bi})$	19.7 minutes	beta, gamma
Polonium-214	$(^{214}\text{Po})$	164 microseconds	alpha
Lead-210	$(^{210}\text{Pb})$	22.3 years	beta, gamma, x-rays
Bismuth-210	$(^{210}\text{Bi})$	5.01 days	beta
Polonium-210	$(^{210}\text{Po})$	138 days	alpha
Lead-206	$(^{206}\text{Pb})$	stable	
<b><u>Thorium-232</u></b>	$(^{232}\text{Th})$	14.1 billion years	alpha, x-rays
Radium-228	$(^{228}\text{Ra})$	5.75 years	beta
Actinium-228	$(^{228}\text{Ac})$	6.13 hours	beta, gamma, x-rays
Thorium-228	$(^{228}\text{Th})$	1.91 years	alpha, gamma, x-rays
Radium-224	$(^{224}\text{Ra})$	3.66 days	alpha, gamma
Radon-220	$(^{220}\text{Rn})$	55.6 seconds	alpha
Polonium-216	$(^{216}\text{Po})$	0.15 seconds	alpha
Lead-212	$(^{212}\text{Pb})$	10.64 hours	beta, gamma, x-rays
Bismuth-212	$(^{212}\text{Bi})$	60.6 minutes	alpha, beta, gamma, x-rays
Polonium-212	$(^{212}\text{Po})$	0.305 microseconds	alpha
Thallium-208	$(^{208}\text{Tl})$	3.07 minutes	beta, gamma
Lead-208	$(^{208}\text{Pb})$	stable	

$^{87}\text{Rb}$  - Drives mantle heat flux.



From Hull (1996a,b; 1997)

Figure 2. A Simplified Decay Scheme of the Uranium Decay Series

**Table 3B. Principal Natural Radionuclide in Non-Decay Series -  $^{40}\text{K}$  and  $^{87}\text{Rb}$** 

Nuclide	Symbol	Half-Life ( $t_{1/2}$ )	Major Radiations
<b>Potassium-40</b>	( $^{40}\text{K}$ )	1.28 billion years	beta, gamma
Argon-40	( $^{40}\text{Ar}$ )	stable	
Calcium-40	( $^{40}\text{Ca}$ )	stable	
<b>Rubidium-87</b>	( $^{87}\text{Rb}$ )	47 billion years	beta
Strontium-87	( $^{87}\text{Sr}$ )	stable	

Source: NRC (1994a)

## GEOLOGY OF NORM

### Igneous Rocks

The original sources of uranium-series, thorium-series, actinium-series, potassium and rubidium radioactivity in the terrestrial environment are the earth's crust and mantle (Table 4).

**Table 4. Crustal Concentrations of Terrestrial Radionuclides**

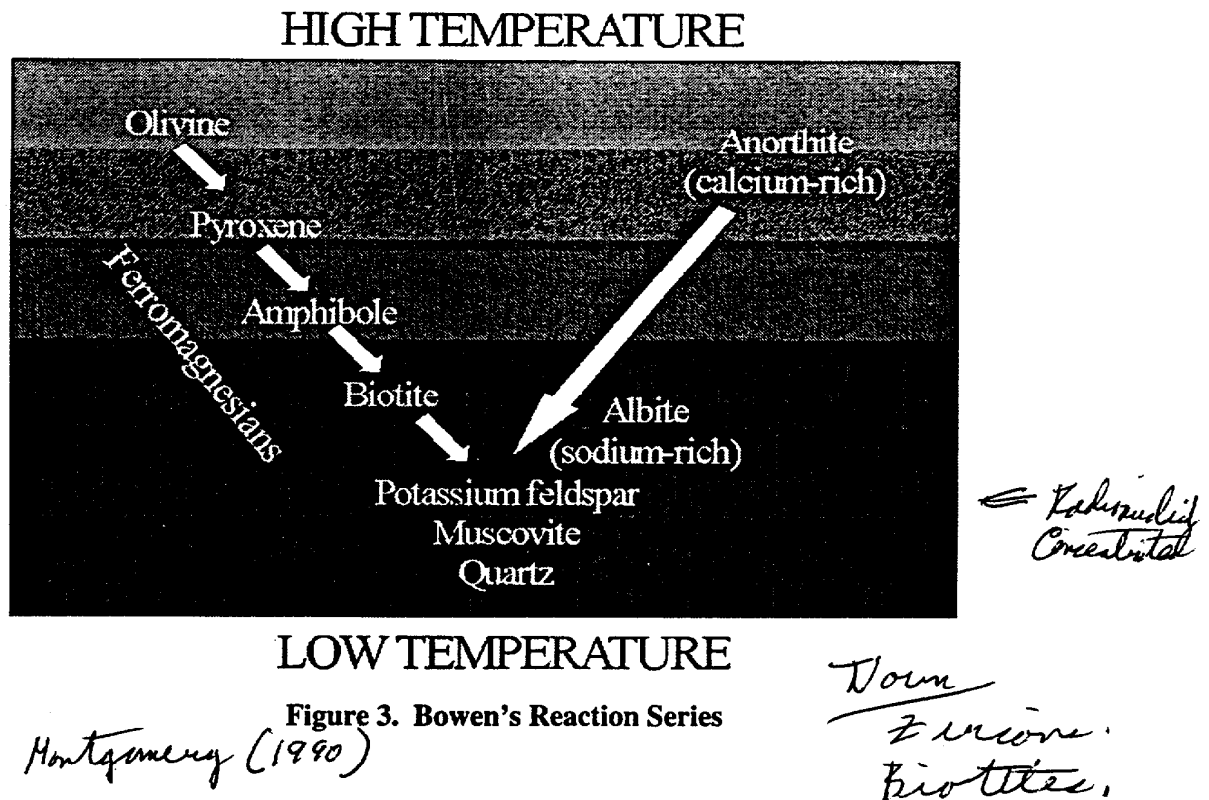
Rock Type	Uranium (U)	Thorium(Th)	Potassium (K)	Rubidium (Rb)
Mafic (Dark Colored)	0.5 to 1 ppm	3 to 4 ppm	0.8%	40 ppm
Salic (Light Colored)	3 ppm	17 ppm	4%	170 to 200 ppm

Source: NCRP 94.

As molten magma cools, silicate minerals crystallize (magmatic differentiation). In the early stages of cooling, the silicates tend to be mafic (those that contain proportionately more iron



and magnesium), and deficient in aluminum, silicon, sodium, and potassium. As cooling and differentiation progress, the silic (containing mostly silicon-aluminum) igneous rocks are formed. Fig. 3 shows a generalization of the process, known as Bowen's Reaction Series (Montgomery 1990). Neither uranium nor thorium "fits" into the crystalline structures of the major silicate minerals so they are excluded by the solid phases and concentrated in the fluid phase of the magma. In addition, they are present in such small quantities as to have little tendency to form minerals in which they would be essential components. The result of this relationship is that the remainder of the magma cools to form varied minor mineral which contain relatively elevated concentrations of uranium, thorium and other minor and trace elements. The last crystallizing silicates contain most of the potassium and rubidium.



### Weathering of Igneous Rocks

Mechanical (physical) and chemical processes break rock down into soil. Weathering plays a key role in this process. Where mechanical processes dominate the breakdown, the separation usually occurs along mineral boundaries that lead to a separation of the major silicates

from the minor ones containing the thorium and uranium. These minor minerals include zircon and monazite. They are stable and resistant to chemical decay and are often found as small individual grains.

Where chemical (or biological) action predominates, the thorium- and uranium-bearing minor minerals can give up their radionuclides to layers of cations in clay minerals. When the host rocks erode, the clay minerals containing with the adsorbed series radionuclides tend to be separated from the major minerals.

Therefore, if igneous rock is broken down to individual grains, the products end up as:

- Sands of the major mineral (depleted of the radionuclides),
- Fine-grained clay minerals (slightly enriched in the radionuclides), and
- Relatively small quantities of resistant, dense grains of the minor minerals containing most of the series radionuclides.
- The potassium and rubidium are removed in solution.

## **Sedimentary Rocks**

Although they comprise only a small part of the earth's crust by volume, sedimentary rocks cover about 85% of the land area of the continental U.S. Therefore, much of the surface soil is derived from sedimentary rock. Sedimentation processes naturally sort the products of weathering and develop several major sedimentary rock types of significantly differing radionuclide concentrations. The major types are:

- Shales
- Sandstones
- Carbonate rocks

As with the igneous rocks, thorium and uranium tend to be minor or disseminated. The radionuclides may become mobile or be deposited by migration of water or oil. Some organic complexes, notable humic acids, create mobile complexes of uranium.

Uranium and other minor and trace elements have an affinity for crude oil. They are probably residues of consolidated organic and marine deposits. Petroleum is often assumed to have migrated to a position of minimum hydraulic potential in a reservoir rock, which may or may not be derived from the same source deposits as the petroleum.

### **Shales**

Shales normally contain at least 35% clay minerals, and a significant fraction contains potassium as an essential constituent. Shales can adsorb the series radionuclides. The radionuclides may also be present bound to organic matter in minor minerals or as precipitates or coprecipitates in the cementing material that binds the rock.

### **Sandstones**

Sandstones are usually made of grains that are primarily quartz, but may contain some potassium-containing feldspars. Those sandstones that contain more than 25% feldspar are called arkoses, and the chief feldspars are those containing potassium. On the whole, sandstones are low in both the series and non-series radionuclides. However, many deposits of uranium are found at the boundary of different layers of sandstones.

### **Carbonate Rocks**

Carbonate rocks are limestones or dolomites derived by chemical precipitation from water or by the buildup of shells, bones, and teeth of organisms. Although the carbonate minerals themselves are relatively free of radionuclides with the exception of U, the intergranular spaces may contain elements found in the seawater from which they were deposited. Potassium is very soluble and does not stay in the deposited matter. Thorium is depleted in seawater and is not metabolized by marine organisms. Therefore, potassium and thorium are usually of low

concentrations in carbonate rocks, but uranium may be present because it may be fixed by reducing conditions in decaying organic matter where the rocks are deposited. Uranium also is chemically speciated as uranyl carbonate aqueous complexes.

Uranium can replace calcium or be adsorbed in the principal phosphate minerals. U is associated with phosphates, as will be discussed in more detail in a following section.

### **Metamorphic Rocks**

The characteristics of metamorphic rocks are based on those of the parent rock in many instances. However, fluids involved in metamorphic processes are often enriched in “eutectic compositions” and these often contain elevated concentrations of U and other NORM. Significant fractionation of decay-series radionuclides can occur during metamorphic events as radioelements are re-distributed in fluids and partial melting zones.

### **Soils**

Radioactivity in soils results from the rock from which it is derived. It is often:

- Diminished by leaching of water,
- Diluted by increased porosity and by added water and organic matter, and
- Augmented by sorption and precipitation of radionuclides from incoming water.

It is the top 0.25 m of soil that contributes significantly to background dose. **Table 5A** is a summary of concentrations of long-lived radionuclides in major rock types and soil. **Table 5B** is a compilation of data for U and Th in accessory mineral phases. Background concentrations of radionuclides in soil vary because of many factors. Soil may have been produced from the weathered top layer of still-intact bedrock below or transported laterally from the same rock unit or type some distance away. Some methods of transport are:

**Table 5A. Concentrations of Long-Lived Radionuclides in Major Rock Types and Soils.**

Rock Type	Potassium-40 <sup>40</sup> K		Rubidium-87 <sup>87</sup> Rb		Thorium-232 <sup>232</sup> Th		Uranium-238 <sup>238</sup> U	
	Percent total K	Bq kg <sup>-1</sup>	ppm of Total Rb	Bq kg <sup>-1</sup>	ppm	Bq kg <sup>-1</sup>	ppm	Bq kg <sup>-1</sup>
<b><i>Igneous Rocks</i></b>								
Basalts - Average	0.8	300	40	30	3-4	10-15	0.5-1	7-10
Mafic Basalts	0.3 - 1.1	7-400	10-50	1-40	1.6, 2.7	7, 10	0.5, 0.9	7-10
Rhyolites	4.5	>1000	170-200	150-180	16, 20	60, 80	3.9, 4.7	50, 60
Granite - Average	>4	>1200	170-200	150-180	17	70	3	40
<b><i>Sedimentary Rocks</i></b>								
Shales -								
Sandstones	2.7	800	120	110	12	50	3.7	40
Quartz Sandstones	<1	<300	<40	<40	<2	<8	<1	<10
Silty Sandstones	2	400	90	80	3-6	10-25	2-3	40
Arkoses	2-3	600-900	80-120	80	2	<8	1-2	10-25
Beach Sands (unconsolidated)	<1	300	40	40	6	25	3	40
Carbonate Rocks	0.3	70	10	8	2	8	2	25
<b><i>Upper Continental Crust (Lithosphere)</i></b>	2.8	850	112	100	10.7	44	2.8	36
<b><i>Soils</i></b>	1.5	400	65	50	9	37	1.8	66

Source: NCRP 1994

**Table 5B. Concentrations of U and Th in Accessory Minerals.**

Accessory Mineral Phases	Total Uranium (ppm)	Total Thorium (ppm)	Th/U
Allanite {accessory	30-700	500-5000	5-10
{pegmatitic	?-100	1000-20000	High
Apatite {accessory	5-150	20-150	1
{coarse aggregate	10-50(?)	50-250(?)	1-5
Epidote	20-50	50-500	2-6
Ilmenite	1-50	-	-
Magnetite (and other opaques)	1-30	0.3-20	
Monazite	500-3000	25000-200000	25-50
Sphene	100-700	100-600	1-2
Zenotime	500-35000	Low	Low
Zircon {accessory	300-3000	100-2500	0.2-1
{pegmatitic	100-6000	50-4000	1

Adapted from Ivanovich and Harmon (1982)

- Natural phenomena such as earthquakes, volcanoes, glaciers and changes in soil composition from flooding.
- Water is the dominant transporting medium. Glacier-derived deposits are common in the Great Lakes area, New England, and Alaska. Outwash erosion products from mountains may produce a soil surface that is more radioactive than the underlying bedrock.
- Wind can be a significant factor, particularly in the Southwest United States.

### **Mobilization and Redeposition of NORM**

(Largely taken from the CRCPD E-4 Committee Report on NORM Contamination and D&D - CRCPD 1994a).

In addition to the geological weathering of rock and soil, NORM concentrations and exposure rates vary because of physical and chemical processes, both natural and anthropogenic. If mobilized, the NORM radioisotopes are available for transport. When radionuclides are dissolved in groundwater, the isotopes tend to travel with the water until redeposition takes place. Airflow serves to transport fine particulates, combustion or volatilization products. Radon, a noble gas, moves in the vapor phase. Radon emanation coefficients will vary depending on the matrix; vitrified products will release less radon than a sandy matrix. We will discuss radon emanation in the sections pertaining to industrial sectors.

Mobilization by human activity can be intentional or unintentional. Two examples are:

- 1) Uranium extraction by *in situ* leaching maximizes solubility of uranium.
- 2) Unintentional mobilization occurs when the element is desirable for its non-radiological properties. Vanadium and uranium were originally mined for their non-radiological properties. Usually, however, the TENORM isotope is mobilized along with other minerals of interest.

Generally, redeposition of NORM involves the same factors as mobilization. Changes in any of the parameters of a stream of material may result in reduced mobility and subsequent redeposition. These processes may also take place preferentially; concentrations of specific minerals may occur. Examples include chlorination of metallic ores as one step in metal production mobilizes radium, which accompanies uranium in the ore. The high solubility of  $\text{RaCl}_2$  relative to other species leads to extraction of radium wherever the parent mineral is exposed to chloride ions. Production of brine or brine-contaminated oil includes dissolved radium as well, since the brine contains chloride ions.

- Low solubility of alkaline earth  $\text{SO}_4^{2-}$  (sulfate) species is also a factor in redeposition of NORM. Movement of sulfuric acid solutions through piping in mineral extraction processes is known to cause precipitation of scale containing high concentrations of radium. Production of water containing sulfate-bearing solutions can also cause precipitation of pipe scale containing elevated concentrations of uranium.
- Groundwater chemistry may change as the water reaches the surface or as it passes through different strata and the dissolved minerals form at the surface. Changes in pH, oxidation state, or chemical equilibrium may result in precipitation of dissolved minerals. This mechanism accounts for the existence of many ore bodies. Addition of alum and softening chemicals in drinking water treatment plants similarly precipitates radium with the other minerals. Uncontrolled discharges from tailings may contain radionuclides (as well as other heavy metals).
- Oxidation-reduction potential can affect solubility. Variation in oxidation state affects solubility since the formation of many chemical complexes depends strongly on oxidation state. For example, water exposed to sulfur-bearing minerals generally exhibits reducing potential, which may alter the oxidation state of other minerals in contact with the water.

- Adsorption depends on the substrate and the specific species in question. Clays are known to adsorb some chemical species preferentially over others; passage of groundwater through a layer of clay may strip out NORM species that would otherwise travel with the water. Adsorption of radon on activated charcoal is an equilibrium process. Desorption can occur if the ambient radon concentration drops; saturation can prevent further radon removal.
- Ion exchange is used to control water chemistry, typically to remove contaminants, soften potable water, or remove radium. Ion exchange does not cause mobilization of radionuclides, but once mobilized in water, any subsequent treatment by ion exchange has clear potential for reconcentration.
- Temperature-dependent variations in solubility result in increased concentrations of radionuclides, together with other elements in geothermal waters. Thermal processes can mobilize radionuclides. Any high-temperature process such as furnaces, kilns, roasters, calcination, and smelting can volatilize lead and polonium. Combustion of coal or lignite volatilizes some isotopes (thorium, uranium, radium, and bismuth). Subsequent redeposition may occur in process equipment, in pollution control equipment, or in the environment. Minerals dissolved in naturally occurring geothermal waters typically plate out as the temperature drops. Deposits of scale containing substantial concentrations of radionuclides may result.
- As the water comes to the surface and the partial vapor pressure drops, radon dissolved in water partitions into the air. In open air, dilution, convection, and diffusion minimize increases in concentration, but in caves or buildings, higher concentrations of radon and its progeny may result. Brines exhibit similar behavior. Oil mixed with brine brings radium with it; as the chemical and physical conditions change in the pipe string and at the well head, the radium precipitates with other minerals and forms scale inside the piping.



- Mechanical reduction in particle size enhances the potential for mobilization of material, such as erosion, movement of alluvial or glacial particles, bulldozers, mining, and construction. In addition, radionuclide concentrations can increase when part of the matrix is removed, leaving radionuclides behind in increased concentrations in the residue. Extraction of phosphate from ore leaves uranium behind in filter cake; bauxite ore contains aluminum, which is extracted leaving “red mud” containing uranium. Coal combustion leaves uranium in ash and slag.
- Suspended materials settle out of material streams as the velocity decreases. Fines settle out where the current is slowest, with successively coarser material settling out in the faster sections. Such gravimetric separation may result in deposits of zircon or monazite sands. Airborne particles exhibit similar behavior, with the coarser material settling closest to the source.

## VARIABILITY OF BACKGROUND RADIATION

Background radiation varies over a range of concentrations and exposure rates from a variety of causes. The magnitude of variation can be significant over a short distance and also can vary in the same place from time-to-time. The background variance can result from natural as well as human activities. Understanding the characteristics of background radioactivity, and the wide range of background values encountered in the field is beneficial when designing and conducting surveys. This is especially important because some of the current regulatory exclusion limits are set at an activity or exposure rate above background (EPA 1980). Proposed clean-up guidance is essentially indistinguishable from background (NRC 1997). Variation due to geology, chemical and physical mobility and deposition, temporal, and human affects should all be considered.

## TEMPORAL VARIABILITY OF BACKGROUNDS

Temporal changes in background radioactivity range occur over a wide range of timeframes; hours to days, months to years, and centuries or more. Short and medium term

changes in background have been measured (Maiello 1997). There are changes in background from terrestrial and cosmic sources that can be summarized as follows (NUREG-1501):

- Cyclic changes on a daily basis are due to changes in radon concentrations in air, which are dependent on the atmosphere (Fig. 4). Early mornings are typically calm, so radon escaping from the ground stays near the surface causing radon levels to rise. During the day, the sun warms the ground and air near it rises, generating a mixing effect that sends the radon (and its progeny) to higher levels in the atmosphere, thus lowering the radon level.
- Diurnal Variation Dose rates rise gradually as soil dries out. Water shields the radiation coming from the NORM in the ground, and dilutes the concentrations of NORM in the soil. After rainfall, the background values drop.

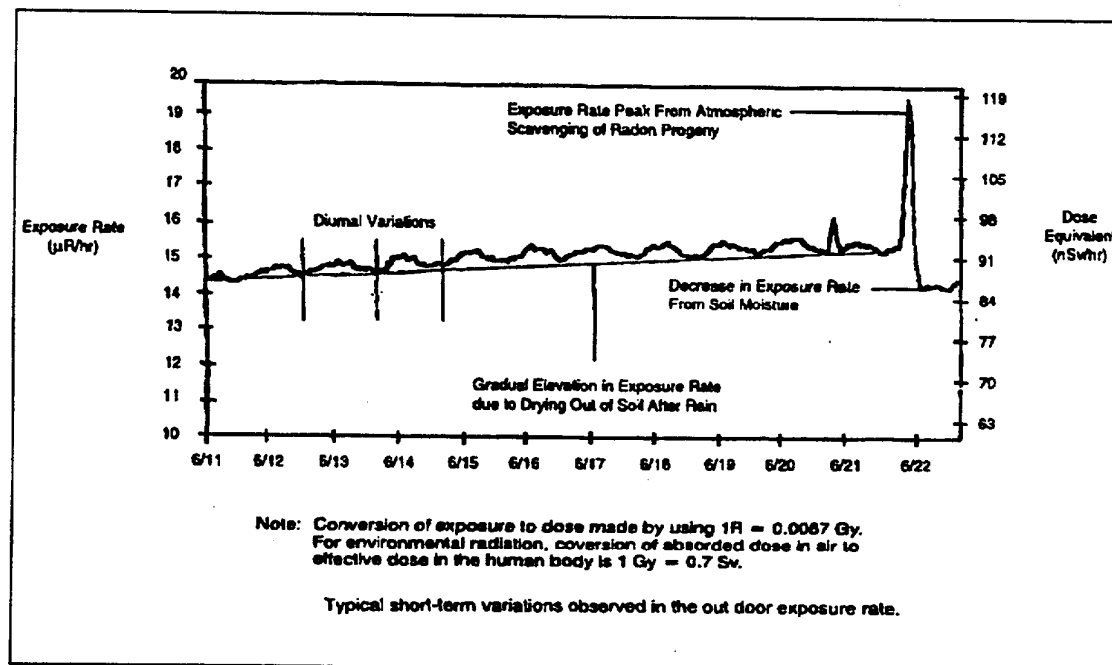
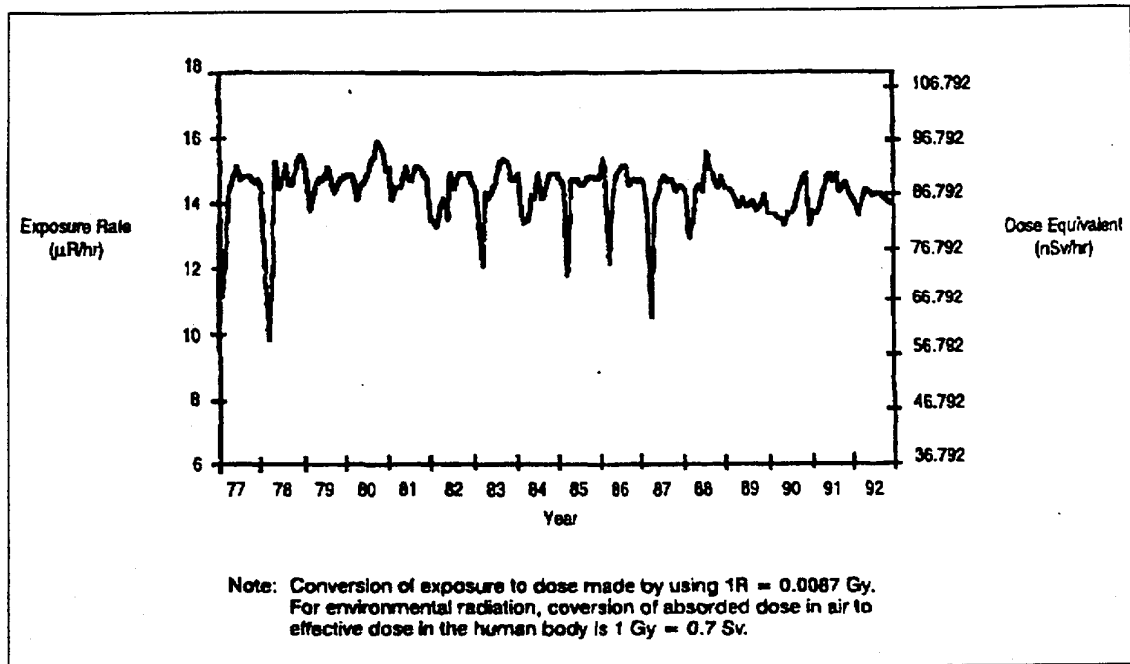
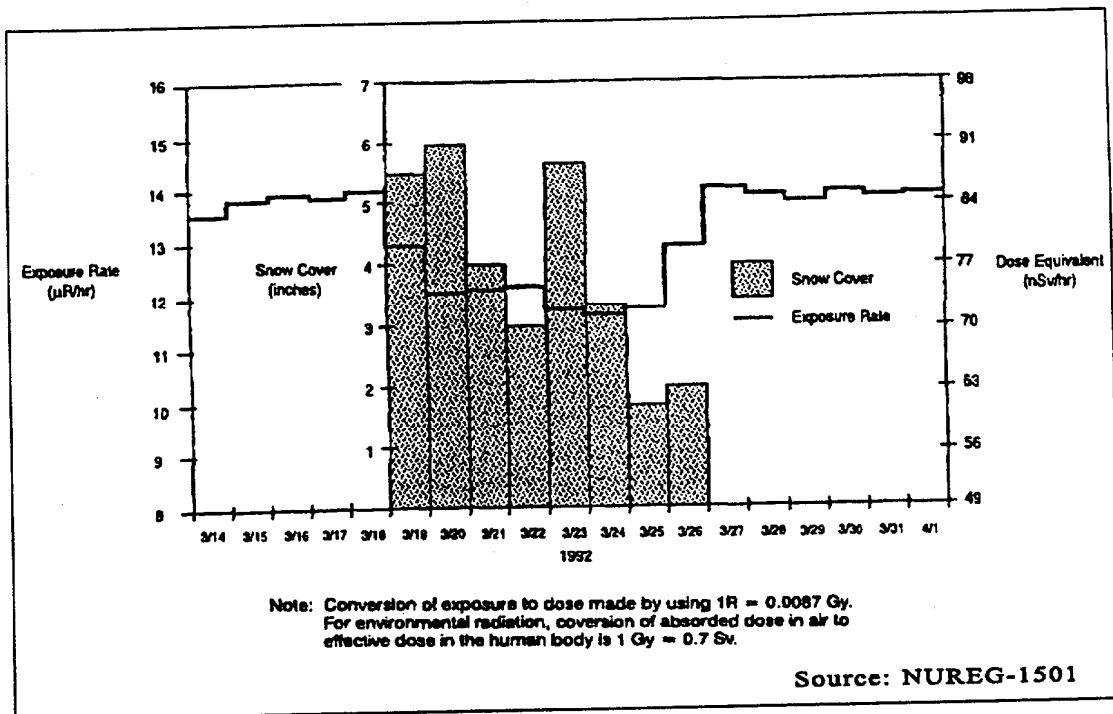


Figure 4. Diurnal Variation of Radon Emanation.



**Figure 5. Shielding effect of snow cover on exposure rates and dose equivalence.**

- Rainfall scours radon and its progeny from the atmosphere, causing radiation levels to rise at ground level. Some larger storms may double the gamma exposure rate for a short period of time.
- The shielding effect of snow is substantial (Fig. 5). Shielding is dependent on the water equivalent of the snow because a heavy wet snow is more effective at shielding than a dry snow.
- Daily and longer term variations in background are due mostly to changes in soil moisture content and snow cover. Winter months trend to lower radiation levels because of higher soil moisture and summer months trend toward higher levels because of lower soil moisture. Seasonal effects actually average out over the course of a year (Fig. 6).



**Figure 6. Daily and Longer Term Variations in Background Radiation.** These variations are primarily due to changes in soil moisture content and snow cover.

- Anthropogenic activities also affect background values. According to NUREG 1501 (NRC 1994a), about two-thirds of the background gamma dose comes from NORM contained in the top 15 cm of soil out to at distance of 6 meters from where a person stands. Therefore, ground coverings, (e.g., asphalt, concrete) may decrease the gamma exposure rate. Conversely, building materials may contribute to dose. The magnitude of change depends on the amount of material involved, and relative radionuclide concentrations in the old and new situations.
- Background increases have been related to nuclear accidents and weapons tests.
- Cosmic ray variations tend to be small and result from changes in barometric pressure. With a high pressure system, there is a larger mass of air to provide a shielding effect, as compared to a low pressure system, which has less air mass and less shielding.

- Cosmogenic radionuclide production varies according to changes in the cosmic ray intensity. A more active sun produces changes in the solar wind and magnetic field, which oppose the cosmic rays coming from outside the solar system.
- Seasonal changes also occur in the deposition of cosmogenic radionuclides on the ground. Deposition is greater during the spring months when air in the stratosphere mixes with air in the troposphere. As mixing occurs, cosmogenic material fluxes to the surface usually increase with higher precipitation rates.

## INDUSTRIAL SECTORS WITH TENORM

Data in this section are taken primarily from EPA (1993).

The majority of TENORM issues center on waste from industrial processes. Most of the wastes that are discussed are produced in very large volumes, but are of relatively low specific radioactivity. While some wastes are disposed of, others are put to commercial uses. The improper disposal, re-use, and recycling of diffuse TENORM has led to circumstances resulting in contamination events and unnecessary public exposures. Disposal in piles or stacks can lead to groundwater contamination and to airborne releases of radioactive particulates and radon. Improper use and/or disposal, such as for soil conditioning or fill around homes, can lead to buildup of radon gas in homes, direct exposure to individuals, and contamination of soil and of the crops growing in the soil. Reuse of TENORM- contaminated materials, such as in concrete aggregate, can lead to increased radiation risks to members of the public in a variety of ways.

This overview is not comprehensive. It is representative of the types of industries that have TENORM. The summaries presented in EPA 1993 were extracted from studies that were conducted to characterize the presence of radioactivity, industry practices, and waste and materials. It should be noted that quality data are not available for many of these industries, and some sectors are not overly willing to share data. Therefore, the data presented in **Table 6** are often extrapolated and best estimates. The main radionuclides investigated in the uranium series

in industrial TENORM situations are  $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{230}\text{Th}$ ,  $^{226}\text{Ra}$ , and  $^{222}\text{Rn}$  (and progeny). In the thorium series, we look at  $^{232}\text{Th}$ ,  $^{228}\text{Ra}$ , and  $^{220}\text{Rn}$  (and progeny). In addition,  $^{40}\text{K}$  should be characterized. Radium-226 is used here to show the relative activity and volume among the TENORM sectors. In assessing dose and risks, all radionuclides need to be considered. Eight industrial sectors will be examined:

- Uranium mining overburden
- Phosphate waste
- Phosphate fertilizers
- Coal ash
- Oil and gas scale and sludge
- Water treatment
- Metal mining and processing (including Rare Earths and Other Metals)
- Geothermal energy production wastes.

*Unregulated By Iode*  
*Po - 210 } More*  
*Pb - 210 }*

*Potash - Pb - Po - 210*

*Host TENORM*  
*→ Phosphate #2*

## Uranium Overburden and Mine Spoils

Unlike ore (source material) and mill tailings (byproduct material), uranium overburden is not regulated by the Atomic Energy Act (AEA 1954) or the Uranium Mill Tailings Remedial Action (UMTRA) program (EPA 1980), and therefore is considered TENORM.

The uranium mining industry began in the 1940s primarily for the purpose of producing uranium ore for use in weapons and soon after for nuclear fuel fabrication. The majority of the mines are located in the west, mainly in Utah, Colorado, Wyoming, Arizona, South Dakota, New Mexico, and Texas. Mining of uranium ores by surface and underground methods produces large amounts of bulk material, including overburden, low-grade ore, and mining spoils. Surface mining produces the bulk of the spoils.

**Table 6. Estimated TENORM Annual Production Rates and Average  $^{226}\text{Ra}$  Specific Activities**

Material/Waste Stream	Production Rate (metric tons <i>per year</i> )	Average $^{226}\text{Ra}$ Concentration, Bq g <sup>-1</sup> (pCi g <sup>-1</sup> )
Uranium overburden	3.8E+07	0.92 (25)
Phosphate waste		
- Phosphogypsum	4.8E+07	1.2 (33)
- Slag	1.6E+06	1.29 (35)
Phosphate fertilizers*	4.8E+06	0.31 (8.3)
Coal ash	6.1E+07	0.14 (3.7)
- Fly ash	4.4E+07	0.14 (3.9)
- Bottom ash and slag	1.7E+07	0.11 (3.1)
Oil and gas scale and sludge	2.6E+06	3.33 (90)
Water treatment	3.0E+06	0.59 (16)
- Sludges	2.6E+06	0.59 (16)
- Radium selective resins	4.0E+04	1295 (35,000)
Metal mining and processing	1.0E+09	0.18 (5)
- Rare Earths	2.1E+03	33.3 (900)
- Zirconium, hafnium, titanium, and tin	4.70E+05	1.59 (43)
- Large volume industries (e.g., copper, iron)	1.0E+09	0.18 (5)
Geothermal energy production wastes	5.4E+04	4.9 (132)

Adapted from EPA (1993) and Hull (1996, 1997)

\*Phosphate fertilizers shown above in **Table 6** are not wastes, but are included in this tabulation due to their widespread use; especially in North America.

Overburden, which overlies the ore deposit, contains limited amounts of natural uranium and its progeny; average  $^{226}\text{Ra}$  concentrations are  $.92 \text{ Bq g}^{-1}$  ( $25 \text{ pCi g}^{-1}$ ) (Table 7).

**Table 7. Radionuclide concentrations of Environmentally Significant Radionuclides in Uranium Mining Overburden**

Radionuclide	Concentration $\text{Bq g}^{-1}$ ( $\text{pCi g}^{-1}$ )
$^{238}\text{U}$	0.92 (25)
$^{234}\text{U}$	0.92 (25)
$^{230}\text{Th}$	0.92 (25)
$^{226}\text{Ra}$	0.92 (25)
$^{210}\text{Pb}$	0.66 (18)
$^{210}\text{Po}$	0.66 (18)
$^{235}\text{U}$	0.048 (1.3)
$^{231}\text{Pa}$	0.048 (1.3)
$^{227}\text{Ac}$	0.048 (1.3)
$^{232}\text{Th}$	0.037 (1.0)
$^{228}\text{Ra}$	0.037 (1.0)
$^{228}\text{Th}$	0.037 (1.0)

Source: EPA (1993)

Mining spoils include low-grade ore and other materials excavated during the mining process. Low-grade ore contains significant amounts of uranium, but usually not enough to make milling economically attractive. The concentrations of  $^{226}\text{Ra}$  at the interface of overburden and low-grade ore boundaries vary from about  $0.1$  to  $\sim 10 \text{ Bq g}^{-1}$  (three to several hundred  $\text{pCi g}^{-1}$ ).



The estimated total volume of mine waste produced is about 4 billion metric tonnes (MT), almost 90% of this within the last 20 years by surface mining. Although demand has fallen off, the deposits that were recently mined are of lower quality and harder to access; therefore, the amount of waste *per* volume of ore generated has increased significantly. As of 1988, there were 3.1 billion MT of unreclaimed overburden in the United States. Uranium mining and milling in America had virtually ceased due to market forces. However, there is now some renewed interest in mining and milling of uranium because stockpiles are low and the price of uranium is rising.

Most uranium overburden is piled and stabilized where it is mined. Uranium overburden has few uses. It is typically used for backfilling mined out areas and for constructing site roads. Mine reclamation will utilize overburden as the practice is implemented. Only about 4% of the mines have been reclaimed. ORNL has found overburden and mine muck used as road aggregate in Colorado (Rice 1995).

Most areas where uranium mining has occurred are remote and arid. These areas are starting to become more populated, and chances for exposures to populations are increasing. A good example of this phenomena is Moab, Utah. The population of Moab was stagnant and actually decreasing during the 1970s and early 1980s. The population of Moab has increased dramatically in recent years with the advent of recreational activities like mountain biking and river rafting. Not only is the population increasing, but tourism in the back country is increasing. The possibility of exposure to TENORM is a concern because numerous uranium mines are located in eastern Utah. These abandoned mines have spoils piles that may not be under any control, and can be accessible. Old mining roads into the back country are used by the recreationists.

Radon concentrations are reduced by escape through diffusion and advection at varying rates. The amount of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  are also reduced by the amount of radon lost. If a radon emanation coefficient of 0.3 is used (sandstone matrix), the  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations are 0.7 times that of  $^{226}\text{Ra}$  or about  $0.66 \text{ Bq g}^{-1}$  ( $18 \text{ pCi g}^{-1}$ ). Radon rates were not given because its release rate from the surface of the overburden was determined from the  $^{226}\text{Ra}$  concentration.

## Phosphate Industry Wastes

Phosphate rock extraction is the fifth largest mining industry in the United States in terms of quantity of material mined. Domestic production from these open pit mines was 38 million metric tonnes (MT) in 1988. Florida produces about 80% of domestic capacity, with North Carolina and Tennessee generating 10% and Idaho, Utah, Montana and Wyoming the balance. Phosphate rock is processed to produce phosphoric acid and elemental phosphorus. These are then combined with other chemicals to produce phosphate fertilizers, detergents, animal feeds, other food products, and phosphorous chemicals. The production of fertilizers accounts for over 90% of the phosphate rock demand in the United States.

Phosphate ore contains one-third quartz sands, one-third clay minerals, and one-third phosphate particles. Uranium in phosphate ores ranges in concentration from 20 to 300 ppm ( $0.26$  to  $3.7 \text{ Bq g}^{-1}$ ) ( $7$  to  $100 \text{ pCi g}^{-1}$ ). Thorium is present in background amounts,  $\sim 1$  to  $5 \text{ ppm}$  ( $3.7$  to  $22.2 \text{ mBq}$ ) ( $0.1$  to  $0.6 \text{ pCi g}^{-1}$ ). When the phosphate particles are separated from the bulk ore (beneficiated), two types of wastes are produced: phosphatic clay tailings and sand tailings (Fig.7). The clay slimes contain 48% of the radionuclides in the host ore, the sand tailings contain 10%, and the remaining 42% are carried by the phosphate rock. Florida clay slime contains about  $1.67 \text{ Bq g}^{-1}$  ( $45 \text{ pCi g}^{-1}$ )  $^{226}\text{Ra}$ .

Phosphogypsum is the principal waste by-product generated during the phosphoric acid production process (wet process), and phosphate slag is the principal waste by-product generated from the production of elemental phosphorous (thermal process). It is estimated that there have been over 8.2 billion MT (9.1 billion short tons) of phosphogypsum generated globally between 1910 and 1991. Impurities contained in the phosphogypsum and phosphate slag include uranium and thorium and their progeny (Table 8). These can become concentrated in the waste by-products.

Phosphatic clay tailings  
Sand Tailings  
Phosphogypsum

Clay Slime 50%  
Sand Tailings 10%

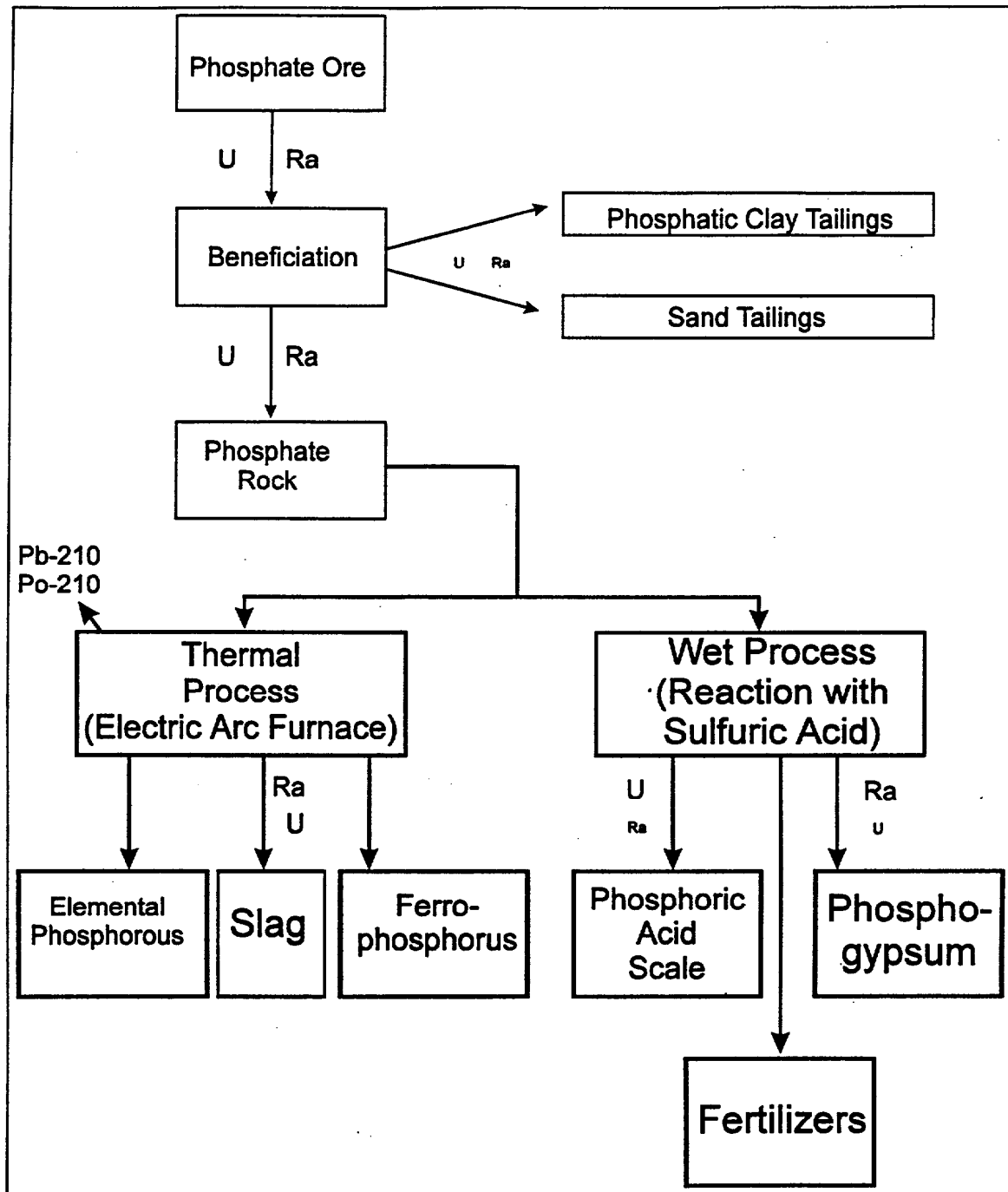
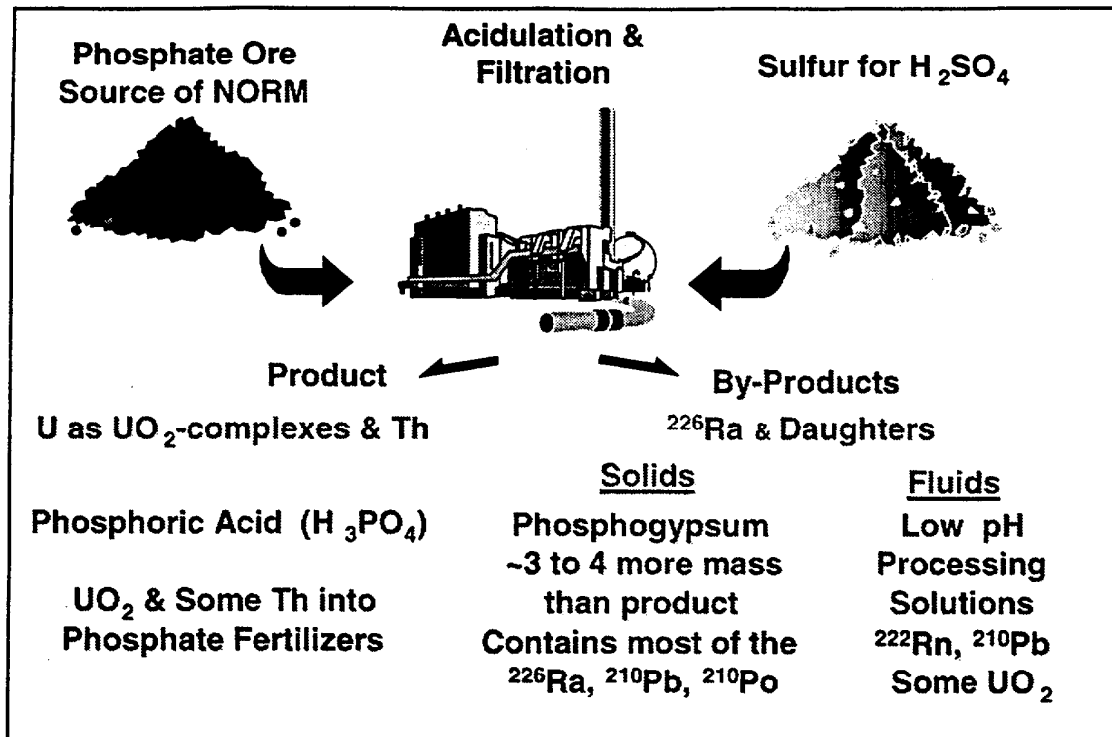


Figure 7. Schematic Production Flow Diagrams of the Thermal and Wet Processes for Manufacture of Phosphate Fertilizers.

**Table 8. Radionuclide Concentrations in Phosphogypsum**

Radionuclide	Concentration, Bq g <sup>-1</sup> (pCi g <sup>-1</sup> )
<sup>238</sup> U	0.22 (6.0)
<sup>234</sup> U	0.23 (6.2)
<sup>230</sup> Th	0.48 (13)
<sup>226</sup> Ra	1.22 (33)
<sup>210</sup> Pb	0.96 (26)
<sup>210</sup> Po	0.96 (26)
<sup>235</sup> U	0.01 (0.30)
<sup>231</sup> Pa	0.01(0.30)
<sup>227</sup> Ac	0.01 (0.30)
<sup>232</sup> Th	0.009 (0.27)
<sup>228</sup> Ra	0.009 (0.27)
<sup>228</sup> Th	0.052 (1.4)

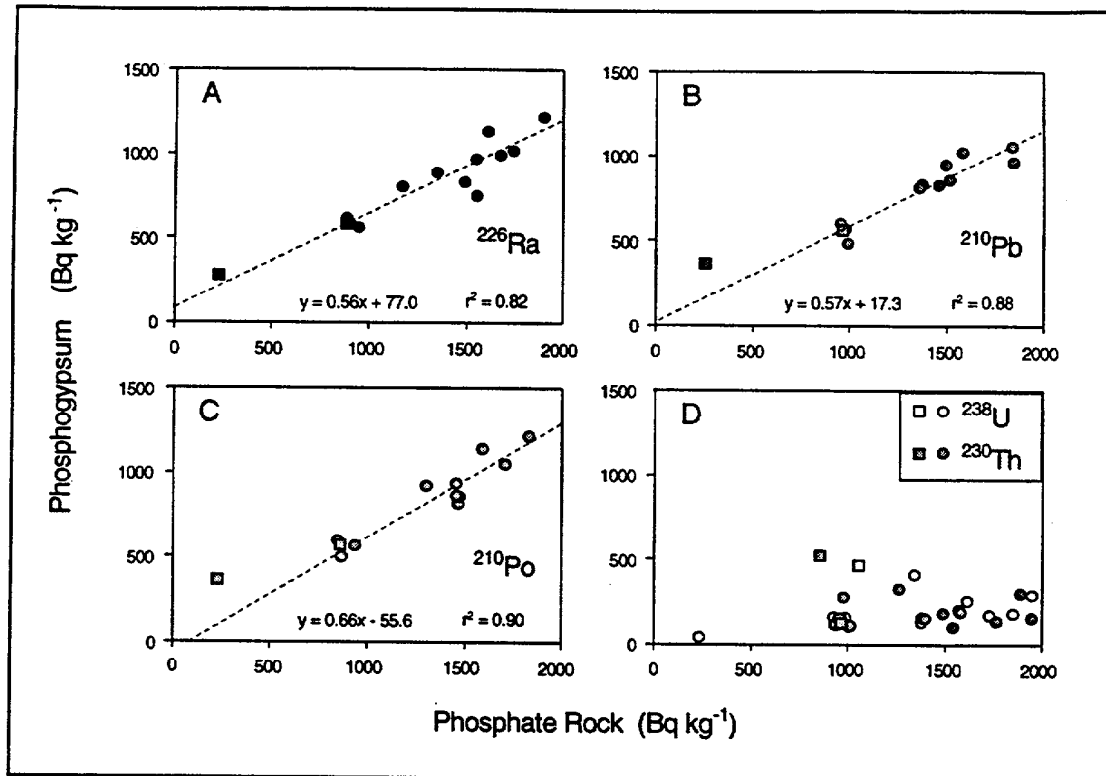
Source: EPA (1993) and Hull (1996a,b; 1997)



**Figure 8. A Generalized Flow-Chart of the "Wet Process" Production of Phosphoric Acid from Phosphate Ore Rock.**

A schematic diagram of the "wet-process" of phosphoric acid production and the fractionation of TENORM in this process is shown in Fig. 8. The sulfur is usually processed into sulfuric acid at the fertilizer production facility. The sulfuric acid, water, and ore are reacted under controlled conditions to maximize the production of phosphoric acid. The phosphoric acid is filtered through large, woven polypropylene filter screens under a partial vacuum to separate the majority of solid, by-product phosphogypsum (PG). The phosphoric acid is pumped to storage tanks and again filtered prior to reaction with liquid ammonium to produce diammonium phosphate fertilizers. These fertilizers contain almost all the uranium and most the thorium from the ore rocks.

About 85% of the  $^{226}Ra$  follows the phosphogypsum, while about 86 % of the uranium and 70% of the thorium are found in the phosphoric acid. Typical radium concentrations in Florida



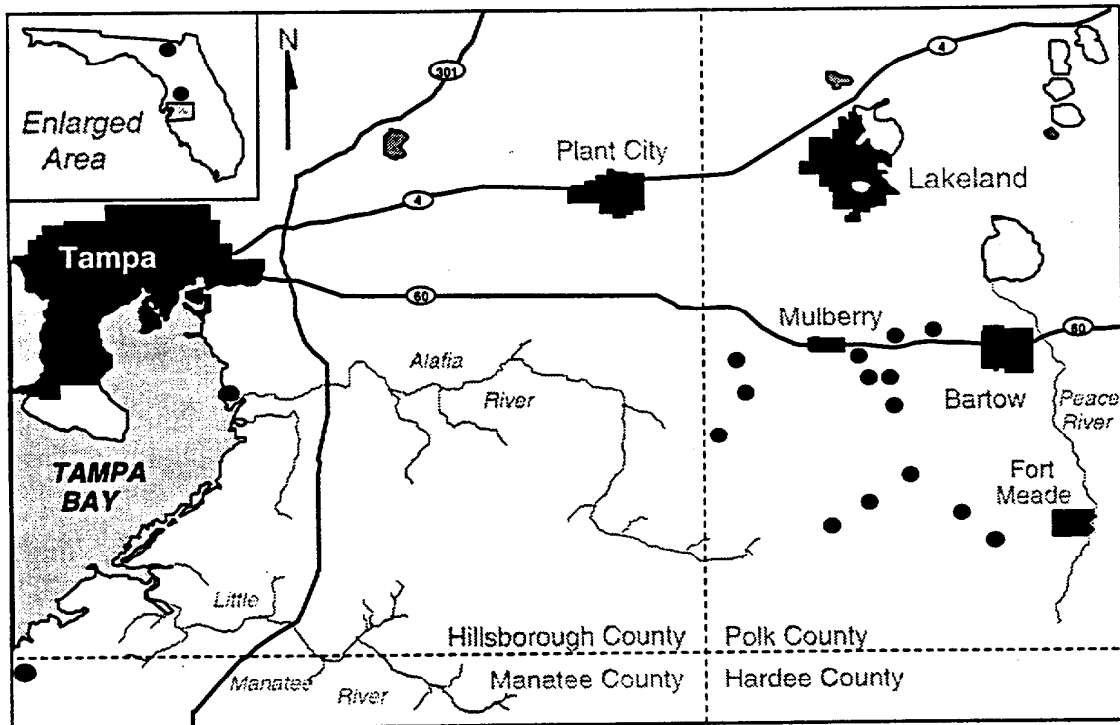
**Figure 9. Cross-Plots of the Fractionation Of <sup>238</sup>U, <sup>230</sup>Th, <sup>226</sup>Ra, <sup>210</sup>Pb, and <sup>210</sup>Po from Phosphate Ore to the Corresponding Phosphogypsum By-Product.**

phosphogypsum stacks fall within a range of 0.41 to 1.3 Bq g<sup>-1</sup> (11 to 35 pCi g<sup>-1</sup>), with progeny also in that range. About 88% to 92% of the radium (<sup>226</sup>Ra) and lead (<sup>210</sup>Pb) are fractionated to the phosphogypsum.

During the wet process, there is selective separation and concentration of radionuclides. Cross-plots of radionuclides in the phosphate ore rock being acidulated (plotted on the abscissa) at the time the corresponding phosphogypsum samples (ordinate) were collected as shown in **Fig. 9**. These plots illustrate the fractionation and re-distribution of radionuclides of interest during the processing of phosphate rock into phosphoric acid in Florida. Virtually all of the polonium (<sup>210</sup>Po) is also included in the phosphogypsum. The slopes of the best-fit, regressed lines for these ore rock - phosphogypsum pairs is not 1 due to the stoichiometry of the reaction; about 1.7 grams

of phosphogypsum are produced for each gram of phosphate rock. It is easily discerned from this graph most of the U and Th are ending up in the phosphoric acid and processing solutions.

Phosphate production wastes, primarily phosphogypsum, is stored in large stacks which are commonly referred to as “gyp-stacks.” The solid by-product PG is slurried with processing solutions and discharged onto the adjacent gyp-stack.. These stacks are huge and often cover 4 to 12 km<sup>2</sup> with an average height of 35 m. The slurries “de-water” and huge volumes of PG accumulate in storage “stacks” that cover a number of square kilometers at each fertilizer plant. Each facility may have one or more stacks that range from 2 to 300 hectares and range in height from 3 to 60 meters. Much of the stack is covered with low pH (2.5 or less), very high ionic strength solutions in ponds and ditches.



**Figure 10. Phosphogypsum storage stacks in Florida (gyp-stacks).** Most stacks are not lined to prevent contamination of the groundwater aquifers with high ionic strength, low pH solutions that contain TENORM.

Within Florida alone there are over 20 active and inactive gyp-stacks (phosphogypsum is no longer being deposited on inactive stacks). The locations of these gyp-stacks in Florida are shown in Fig. 10. Within the next two years storage of this water-soluble, radioactive waste shall exceed  $1 \times 10^9$  tons. Most of these “gyp-stacks” have been deposited directly on the land surface without any liners or other physical to reduce the flux of high ionic strength and radionuclide-bearing solutions into groundwater reservoirs. One of the larger of these “gyp-stacks” recently “fell-into” the Floridan Aquifer system. The acidic solutions emanating from these stacks dissolved the underlying carbonate rocks. Thousands of tons of this material was introduced into one of the largest fresh water aquifer systems in North America just upgradient of Tampa, Florida.

Radon flux rates from phosphogypsum stacks vary widely, due to the radium concentration in the parent rock, the emanation fraction, and other factors. Average fluxes have been reported to vary from  $0.063$  to  $0.44 \text{ Bq m}^{-2} \text{ s}^{-1}$  ( $1.7$  to  $12 \text{ pCi m}^{-2} \text{ s}^{-1}$ ), with a mean value of  $0.25 \text{ Bq m}^{-2} \text{ s}^{-1}$  ( $6.8 \text{ pCi m}^{-2} \text{ s}^{-1}$ ). The radon emanation coefficient for phosphogypsum is estimated at a value of 0.2.

Gamma radiation exposure rates from phosphogypsum stacks have been measured around  $\sim 0.287 \text{ } \mu\text{Sv hr}^{-1}$  ( $33 \text{ } \mu\text{R hr}^{-1}$ )<sup>1</sup>. Radiation surveys conducted in areas where large volumes of phosphate ores are stockpiled have yielded gamma exposure rates ranging from  $\sim 0.174$  to  $0.87 \text{ } \mu\text{Sv hr}^{-1}$  ( $20$  to  $100 \text{ } \mu\text{R hr}^{-1}$ ), with an average of  $\sim 0.522 \text{ } \mu\text{Sv hr}^{-1}$  ( $60 \text{ } \mu\text{R hr}^{-1}$ ).

Some phosphogypsum is used for agricultural and construction purposes. EPA has ruled that “Phosphogypsum intended for agricultural use must have a certified average concentration of  $^{226}\text{Ra}$  of no greater than  $10 \text{ pCi g}^{-1}$ ” (EPA 1992). However, virtually no phosphogypsum has specific activities of  $^{226}\text{Ra}$  in this low range; the great majority averages  $20$  to  $35 \text{ pCi g}^{-1}$  (Hull, 1997).

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<sup>1</sup> Conversion from exposure to dose made by using  $1\text{R}=0.0087 \text{ Gy}$ . A quality factor of 1.0 is used to convert from Gy to Sv.



In contrast to the “wet-process,” during the thermal process vitrification yields slag, a material that contains the non-volatile radionuclides. This slag has been found to contain uranium and thorium concentrations in the range of 0.74 to 1.85 Bq g<sup>-1</sup> (20 to 50 pCi g<sup>-1</sup>) and <sup>226</sup>Ra concentrations in the range of 0.15 to 1.5 Bq g<sup>-1</sup> (4 to 40 pCi g<sup>-1</sup>) (Table 9). Because of the high temperatures, some radionuclides are vaporized during the process. As much as 95% of the <sup>210</sup>Pb and <sup>210</sup>Po have been measured in stack releases. Eventually, these isotopes decay and grow back into equilibrium with the <sup>226</sup>Ra.

**Table 9. Radionuclide Concentrations in Phosphate Slag**

Radionuclide	Concentration Bq g <sup>-1</sup> (pCi g <sup>-1</sup> )
<sup>238</sup> U	0.92 (25)
<sup>234</sup> U	0.88 (24)
<sup>230</sup> Th	1.19 (32)
<sup>226</sup> Ra	1.26 (35)
<sup>210</sup> Pb	1.26 (35)
<sup>210</sup> Po	1.26 (35)
<sup>235</sup> U	0.05 (1.3)
<sup>231</sup> Pa	0.05 (1.3)
<sup>227</sup> Ac	0.05 (1.3)
<sup>232</sup> Th	0.03 (0.77)
<sup>228</sup> Ra	0.03 (0.77)
<sup>228</sup> Th	0.03 (0.77)

Source: EPA (1993)

The total slag inventory in the United States in 1991 is estimated at 224 to 424 million MT (247 to 467 million short tons). The radon emanation coefficient for slag is estimated to be very low because of the vitrified matrix. A value of 0.01 was assumed for the referenced report.

Radon flux measurements conducted on Idaho slag indicate that very little radon escapes the vitrified slag matrix. An average radon flux rate of  $0.02 \text{ Bq m}^{-2}\text{sec}^{-1}$  ( $0.5 \text{ pCi m}^{-2} \text{ sec}^{-1}$ ) is estimated for a typical phosphate slag pile. For comparison, measurements taken on two phosphate ore samples revealed radon fluxes of  $2.11$  and  $2.37 \text{ Bq m}^{-2}\text{sec}^{-1}$  ( $57$  and  $64 \text{ pCi m}^{-2} \text{ sec}^{-1}$ ); radon fluxes from native soil samples ranged from  $0.063$  to  $0.63 \text{ Bq m}^{-2}\text{sec}^{-1}$  ( $1.7$  to  $17 \text{ pCi m}^{-2} \text{ sec}^{-1}$ ).

Gamma radiation exposure rates of  $\sim 0.87 \mu\text{Sv hr}^{-1}$  ( $100 \mu\text{R hr}^{-1}$ ) have been measured on slag piles. Phosphate slag has been used as aggregate in making roads, streets, pavements, residential structures, concrete aggregate, railroad ballast, and buildings. Radiation surveys conducted in Montana and Idaho where slag has been used in construction materials and to pave streets have yielded measurements of  $\sim 0.565 \mu\text{Sv hr}^{-1}$  ( $65 \mu\text{R hr}^{-1}$ ) in homes and  $\sim 0.435 \mu\text{Sv hr}^{-1}$  ( $50 \mu\text{R hr}^{-1}$ ) on streets that utilized slag.

### **Phosphate Fertilizers and Potash**

Phosphate fertilizers are one of the end products from the phosphate industry just discussed. Phosphate and potassium are also found in multiple-nutrient fertilizers, which are available in different blends of nitrogen (N), phosphorous (P), and potassium (K).

Potash is another material used as a fertilizer that contains natural radioactivity, primarily  $^{40}\text{K}$ . Potash is composed principally of the salts of potassium, of which potassium chloride and potassium sulfate are the major components.

Phosphate fertilizers are produced by mixing phosphoric acid directly with phosphate rock. Ammonia and potassium salts are also added to produce a variety of fertilizers. Mined from sylvinite ore or produced by solar evaporation, potash can be used directly as a fertilizer without extensive chemical conversion. The continued widespread use of phosphate fertilizers may eventually result in a measurable increase in background radiation levels.

Radionuclide concentrations vary with the type of fertilizer and production process, with average concentrations ranging from 0.18 to 0.74 Bq g<sup>-1</sup> (5 to 20 pCi g<sup>-1</sup>) for <sup>226</sup>Ra, 0.74 to 2.22 Bq g<sup>-1</sup> (20 to 60 pCi g<sup>-1</sup>) for uranium, and 0.037 to 0.18 Bq g<sup>-1</sup> (1 to 5 pCi g<sup>-1</sup>) for thorium (Table 10). The activity of <sup>40</sup>K in potash depends of the quantity of potassium present, which is normally expressed as equivalent mass of K<sub>2</sub>O. The equivalent concentration of <sup>40</sup>K in potash is about 25.75 Bq g<sup>-1</sup> (696 pCi g<sup>-1</sup>) K<sub>2</sub>O. Since marketable potash contains about 60% K<sub>2</sub>O, the concentration of <sup>40</sup>K in the final product calculates to approximately 15.5 Bq g<sup>-1</sup> (420 pCi g<sup>-1</sup>).

Radon fluxes for phosphate fertilizers in soil are expected to be similar to those for unfertilized soils. A typical flux for a fertilized soil is approximately 0.037 Bq m<sup>-2</sup> (1.0 pCi m<sup>-2</sup>) per pCi g<sup>-1</sup> of <sup>226</sup>Ra. The external gamma radiation attributable to fertilizer materials is only about 0.25% of that from unfertilized soil.

**Table 10. Radionuclide Concentrations in the Average Phosphatic Fertilizer**

Radionuclide	Phosphate Fertilizer	Potash
	Concentration, Bq g <sup>-1</sup> (pCi g <sup>-1</sup> )	
<sup>40</sup> K	-	25.75 (696)
<sup>238</sup> U	2.04 (55)	-
<sup>234</sup> U	2.07 (56)	-
<sup>230</sup> Th	1.96 (53)	-
<sup>226</sup> Ra	0.31 (8.3)	-
<sup>210</sup> Pb	0.22 (5.8)	-
<sup>210</sup> Po	0.22 (5.8)	-

$^{235}\text{U}$	0.096 (2.6)	-
$^{231}\text{Pa}$	0.096 (2.6)	-
$^{227}\text{Ac}$	0.096 (2.6)	-
$^{232}\text{Th}$	0.037 (1.0)	-
$^{228}\text{Ra}$	0.037 (1.0)	-
$^{228}\text{Th}$	0.037 (1.0)	-

Source: (EPA 1993)

### Coal Ash

There are over 1,300 coal-fired boilers operated by electric utilities and nearly 60,000 industrial boilers in the United States. Electric utilities consume the most coal, currently at about 700 million MT (771 million short tons) *per year*. Domestic coal production has increased, as well as imports, while exports have remained relatively stable. In 1990, 61.6 million MT (67.9 million short tons) of ash and slag were generated, with another 17.2 million MT (18.9 million short tons) of sludges. Coal consumption generates large amounts of coal ash that requires proper management and disposal, either at the point of use or elsewhere in ash impoundment facilities. Since coal contains naturally occurring uranium and thorium, large quantities of coal ash may present a potential radiological risk to exposed individuals. The degree of risk will depend on the physical and radiological properties of the ash and on how the ash is disposed of or used.

The radioactivity of coal can vary over two orders of magnitude depending on the type of coal and the region from which it is mined. The concentrations of  $^{238}\text{U}$  and  $^{232}\text{Th}$  in coal average about 0.022 and 0.018 Bq g<sup>-1</sup> (0.6 and 0.5 pCi g<sup>-1</sup>), respectively. The concentrations of the radionuclides in ash will also vary (Table 11). They tend to be enriched in ash compared to coal.

Electrical utility boilers generate ash at a rate of about 10% of the original volumes of coal. Over 95% of the ash is retained. Bottom ash and slag make up about 20% and fly ash makes up the other 75%. Fly ash is formed when flue gases entrain (to draw after oneself) ash.

Fly ash is very fine. The remainder of the ash that is too heavy to go off with the gas settles to the bottom of the boiler to become bottom ash.

Ash also typically contains silicon, aluminum, iron and calcium. Liquid slag is produced when the ash melts under intense heat. Treatment of stack exhausts also results in the generation of flue gas desulfurization sludges. About 17 million MT (18.75 million short tons) were produced in 1990.

**Table 11. Typical Average Radionuclide Concentrations for Coal Ash**

Radionuclide	Concentration, Bq g <sup>-1</sup> (pCi g <sup>-1</sup> )
<sup>238</sup> U	0.12 (3.3)
<sup>234</sup> U	0.12 (3.3)
<sup>230</sup> Th	0.085 (2.3)
<sup>226</sup> Ra	0.14 (3.7)
<sup>210</sup> Pb	0.25 (6.8)
<sup>210</sup> Po	0.26 (7.0)
<sup>235</sup> U	0.0037 (0.1)
<sup>231</sup> Pa	0.0059 (0.16)
<sup>227</sup> Ac	0.0059 (0.16)
<sup>232</sup> Th	0.077 (2.1)
<sup>228</sup> Ra	0.066 (1.8)
<sup>228</sup> Th	0.19 (3.2)

Source: EPA (1993)

The radon emanation coefficient for ash is low because the ash is vitrified. A factor of 0.02 can be used to compare to other coefficients. Radon flux is also low, estimated at  $0.018 \text{ Bq m}^{-2}\text{sec}^{-1}$  ( $0.5 \text{ pCi m}^{-2} \text{ sec}^{-1}$ ).

About 70 to 80% of the coal ash generated is disposed of in landfills or ponds. There are about 300 off-site coal-ash landfills and surface impoundments. A typical ash disposal landfill may be anywhere from 30 to 60 hectares. It is estimated there are 305 off-site coal-ash landfills and surface impoundments and that there are about 900 on-site disposal facilities. Fly ash, bottom ash, and boiler slags are used as substitutes in cement and concrete, as structural fills, for snow and ice control, and as blasting grits. The potential impact of long-term accumulation of by-products in the biosphere should be considered (Gabbard 1993).

Coal ash is used as an additive in concrete, cement, and roofing materials, land reclamation, paint and undercoatings, and various products and as a structural fill for road construction. About 30% of ash is reused. There is concern that fly ash may become regulated in the future, which would discourage reuse.

### **Oil and Gas Production Scale and Sludge**

The rate of production of domestic crude oil is closely tied to the international price of crude and to fluctuations that depend on world-wide political and economic conditions. Production for the month of November 1995 was estimated at 6.5 million barrels *per day* (API 1996). Production in 1970 was approximately 9.6 million barrels *per day*. It is estimated that about 25 thousand MT (27.5 thousand short tons) of TENORM scale and 230 thousand MT (253.5 thousand short tons) of TENORM sludge are generated from domestic production each year, based on 1989 figures.

Radioactivity in oil and gas production and processing equipment is of natural origin and is now known to be widespread, occurring throughout the world. Estimates suggest that up to

30% of domestic oil and gas wells may produce some elevated TENORM contamination. The geographic areas with the highest recorded measurements were northern Texas and the gulf coast crescent from southern Louisiana and Mississippi to the Florida panhandle. Very low levels of TENORM radioactivity were noted in California, Utah, Wyoming, Colorado, and northern Kansas fields.

Uranium and thorium compounds are mostly insoluble and as oil and gas are brought to the surface, remain in the underground reservoir. As the natural pressure within the bearing formation falls, formation water present in the reservoir will also be extracted with the oil and gas. Some radium and radium daughter compounds are slightly soluble in water and may become mobilized when this production water is brought to the surface. The precipitate consists principally of barium sulfate ( $\text{BaSO}_4$ ), calcium sulfate ( $\text{CaSO}_4$ ), and calcium carbonate ( $\text{CaCO}_3$ ). Because the chemistry of radium is similar to that of barium and calcium (all are Group IIA elements), radium may also precipitate to form complex sulfates and carbonates.

The amount of TENORM material from a producing field generally increases as the amount of water pumped from the formation increases. Since radium concentrations in the original formation are highly variable, the concentrations that precipitate out in sludges and as scale on internal surfaces of oil and gas production and processing equipment are also variable. This scale in these chemical matrices is relatively insoluble and may vary in thickness from a few millimeters to more than an inch. Scale deposits in production equipment may at times become so thick to completely block the flow in pipes as large as 10.1 cm (4 in.) in diameter.

Radium-226 in scales generally has higher specific activities than  $^{228}\text{Ra}$ . Typically,  $^{226}\text{Ra}$  in scale is in equilibrium with its progeny, but  $^{228}\text{Ra}$  is not. The nominal activity appears to be about three times greater for  $^{226}\text{Ra}$  than for  $^{228}\text{Ra}$  (Table 12).

Table 12. Average Radionuclide Concentrations in Oil and Gas Scale

Radionuclide	Concentration, Bq g ( $\mu\text{Ci g}^{-1}$ )
$^{226}\text{Ra}$	13.3 (360)
$^{210}\text{Pb}$	13.3 (360)
$^{210}\text{Po}$	13.3 (360)
$^{228}\text{Ra}$	4.44 (120)
$^{228}\text{Th}$	4.44 (120)

Source: EPA (1993)

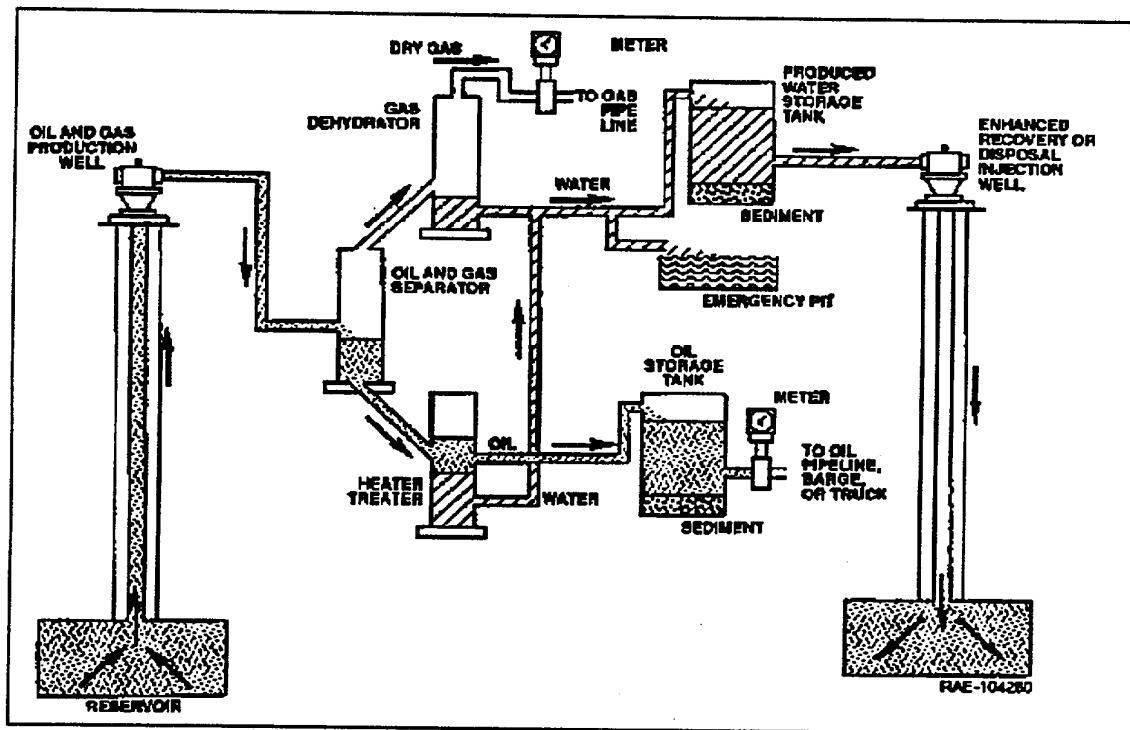


Figure 11. Schematic of Oil and Gas Processing Equipment.



The oil and gas production stream passes through a separator where the oil, gas, and water are divided into separate streams based on their different fluid densities (Fig. 11). Most of the solids removed in the separator accumulate there. The product may also be treated using a heater/treater to separate oil from produced water and sludge. The produced water flows from the separators into storage tanks and is often injected into disposal or recovery wells. Scales are usually found in piping and tubing, including oil flow lines, water lines, injection and production well tubing, manifold piping and small-diameter valves, meters, screens, and filters. The highest concentrations of TENORM occurred in wellhead piping and production piping near the wellhead. Concentrations of radium in scale deposited in production tubing near wellheads can range up to tens of thousands of picocuries *per* gram. The concentration of radium deposited in separators is about a factor of ten less than that found in wellhead systems. There is a further reduction of up to an order of magnitude in the radium concentration in heater/treaters. The activities in granular deposits found in separators range from one to about one thousand picocuries *per* gram. The largest volumes of scale have been found in the water lines associated with separators, heater/treaters, and gas dehydrators.

TENORM radionuclides may also accumulate in gas plant equipment from  $^{222}\text{Rn}$  decay products, even though the gas is removed from its  $^{226}\text{Ra}$  parent. Rn-222 concentrates in the liquid petroleum gas (LPG) fraction during processing. Gas plant deposits differ from oil production scales, typically consisting of radon decay products plated out on the interior surfaces of pipes, valves, and other gas plant equipment. The only significant radionuclides remaining in gas plant equipment are  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ .

Radon flux rates from scale are hard to determine. Several factors, such as particle size, thickness of the deposit, and the presence of oil and other material may reduce radon flux rates. Since much of the waste is internal to components, it may be challenging to characterize net radon flux. A 0.05 radon emanation coefficient has been assumed for the referenced report (EPA, 1993).

Exposure rates vary widely depending on geographic location and the type of equipment. Median exposure rates were measured for water handling equipment in the  $\sim 0.261$  to  $0.348 \mu\text{Sv hr}^{-1}$  (30 to  $40 \mu\text{R hr}^{-1}$ ) range. Gas processing equipment with the highest levels include reflux

pumps, propane pumps, and tanks and lines. Median exposure rates were reported to be in the  $\sim 0.348$  to  $0.609 \mu\text{Sv hr}^{-1}$  ( $30$  to  $70 \mu\text{R hr}^{-1}$ ) range. For both oil and gas processing equipment, a few measurements were observed to be in excess of  $\sim 8.7 \mu\text{Sv hr}^{-1}$  ( $1 \text{ mR hr}^{-1}$ ).

The origin of TENORM-contaminated sludge is similar to that of scale. As the produced water is subjected to changes in temperature and pressure, dissolved solids may precipitate out of solution and deposit sludge within the oil production system. These deposits are generally in the form of oily, loose material. Sludge often contains silica compounds, but may also contain significant amounts of barium. Some of the solids in the original product stream are removed in the separator and accumulate there as sludge. As the stream is further treated using heater/treaters to separate oil from water, sludge is also separated and allowed to accumulate. The largest volumes of sludge settle out of the production stream and remain in the oil stock and water storage tanks. Radionuclide concentrations in sludge vary from background levels to several hundred picocuries *per* gram, with the highest concentrations in the separator and collection areas near the separator (drains, *etc.*) (Table 13). The levels deposited in heater/treaters and in sludge holding tanks are about a factor of 10 less than those found in the separator. TENORM concentrations in sludge deposits in heater/treaters and tanks are generally around  $2.78 \text{ Bq g}^{-1}$  ( $75 \text{ pCi g}^{-1}$ ).

**Table 13. Average Radionuclide Concentrations in Sludge**

Radionuclide	Concentration, $\text{Bq g}^{-1}$ ( $\text{pCi g}^{-1}$ )
$^{226}\text{Ra}$	2.07 (56)
$^{210}\text{Pb}$	2.07 (56)
$^{210}\text{Po}$	2.07 (56)
$^{228}\text{Ra}$	0.7 (19)
$^{228}\text{Th}$	0.7 (19)

Source: EPA (1993)

Radon flux from sludge is also hard to characterize for several reasons. The presence of oil or other petroleum products associated with the sludge may reduce radon flux rates. The presence and concentration of  $^{226}\text{Ra}$  will govern radon flux and diffusion properties from sludge. A radon emanation coefficient of 0.22 was assumed for the referenced report (EPA 1993).

Oil field tubulars and equipment are now surveyed for the presence of radioactivity, and contaminated equipment is either held in storage or sent to a commercial decontamination facility. Tank sludges are also surveyed for radioactivity, dewatered, and held in storage pending disposal.

In some states and provinces, production water from oil and gas industry is disposed down hole. In addition, well injection for slurried material at limited concentrations has been permitted for oil field TENORM. Some oil field scale is stored in drums. The industry disposes of scale and sludge wastes removed from production equipment and also discards contaminated components. There are instances where TENORM waste is disposed of off-shore, under license from the United States Mineral Management Service.

### **Waste Water Treatment Sludge**

Since water for domestic use comes from streams, lakes, reservoirs, and aquifers, it contains varying amounts of naturally occurring radioactivity. Radionuclides are leached into ground or surface water when water comes in contact with uranium- and thorium- bearing geologic media. The predominant radionuclides found in water include radium, uranium, and radon, as well as their progeny.

Water treatment includes passing the water through various types of filters and devices that rely on physical and chemical processes to remove impurities and organisms. If water containing radionuclides is treated by such systems, it is possible to generate radioactive wastes even if the treatment system was not originally intended to remove radioactivity. Such wastes include filter sludges, ion-exchange resins, granular activated carbon, and water from filter backwash.

Of the over 60,000 public water supply systems, it was estimated that about 700 of them treat water containing elevated NORM radionuclide concentrations. The areas suspected of having the most systems with elevated radionuclide concentrations are the North Central Region, the Piedmont and Coastal Plain Provinces, and portions of Arizona, New Mexico, Texas, Mississippi, Florida, and Massachusetts.

It is estimated that approximately 260,000 MT (287,000 short tons) of water treatment sludge containing elevated levels of TENORM, including spent resins and charcoal, are generated annually (Table 14).

Three technologies are likely to produce the TENORM waste because they generate sludge and are known to remove radioactivity from water. They are lime softening, greensand filtration, and ion-exchange and activated charcoal.

- Lime softening is used on larger systems to soften water by the addition of calcium hydroxide, which raises the pH causing calcium and magnesium in the water to precipitate. The precipitate, along with the suspended solids, is removed by sedimentation and filtration. Eighty to 90 % of the radium in the water is also trapped in the sludge.
- Greensand is made of grains of glauconite often mingled with clay or sand and may also contain natural algae. These large sand bed filtration systems remove nearly 60% of radium found in the water.
- Ion-exchange resins are used to soften water. Cation exchange removes about 95% of the radium. Anion exchange removes about 95% of the uranium. These resins are usually back- washed for reuse. The backwash water is typically discharged or back-washed to another column for further treatment. Radionuclide content eventually builds up in the resin after prolonged use. Activated charcoal is often used in conjunction with ion-exchange systems to

remove organics and gases, including radon. Over 95% of the radon, with smaller amounts of uranium and radium can be removed.

**Table 14. Average Radionuclide Concentrations in Water Treatment Sludge**

Radionuclide	Influent Water, Bq/L (pCi/L) (above normal concentrations)	Sludge, Bq g <sup>-1</sup> (pCi g <sup>-1</sup> )
<sup>238</sup> U	0.074 (2.0)	0.15 (4.0)
<sup>234</sup> U	0.074 (2.0)	0.15 (4.0)
<sup>230</sup> Th	0.0037 (0.1)	0.0074 (0.2)
<sup>226</sup> Ra	0.30 (8.0)	0.59 (16.0)
<sup>210</sup> Pb	0.18 (4.8)	0.41 (11.0)
<sup>210</sup> Po	0.18 (4.8)	0.41 (11.0)
<sup>235</sup> U	0.00052 (0.014)	0.0011 (0.03)
<sup>231</sup> Pa	0.00052 (0.014)	0.0011 (0.03)
<sup>227</sup> Ac	0.00052 (0.014)	0.0011 (0.03)
<sup>232</sup> Th	0.0037 (0.1)	0.0074 (0.2)
<sup>228</sup> Ra	0.37 (10.0)	0.74 (20.0) [0.59 (16)] <sup>a</sup>
<sup>228</sup> Th	0.0037 (0.1)	0.0074 (0.2) [0.33 (9.0)] <sup>a</sup>

<sup>a</sup> For <sup>228</sup>Ra and <sup>228</sup>Th, the values shown in brackets are concentrations after two years of decay and ingrowth. Adapted from EPA (1993).

Ion-exchange resins generate waste at higher concentrations of those found in sludges but in much smaller quantities. Field data indicate that radium concentrations between 11.8 to 129.5 Bq L<sup>-1</sup> (320 to 3,500 pCi L<sup>-1</sup>) occur in the column rinse and brine. Radium buildup in cation-exchange resins has been observed to average about 0.33 Bq g<sup>-1</sup> (9 pCi g<sup>-1</sup>), with peak concentrations ranging from 0.92 to 1.48 Bq g<sup>-1</sup> (25 to 40 pCi g<sup>-1</sup>).

Selective sorbants specifically designed to remove radium from water are particularly effective, with wastes retaining concentrations of <sup>226</sup>Ra averaging 1.48 kBq g<sup>-1</sup> (40,000 pCi g<sup>-1</sup>) and up to 4.07 kBq g<sup>-1</sup> (110,000 pCi g<sup>-1</sup>). This material is considered discrete NARM, > 74 Bq g<sup>-1</sup> (> 2 nCi g<sup>-1</sup>), and should be treated as low-level waste.

The concentration of radionuclides in water treatment sludge will depend on:

- The amount of naturally occurring radioactivity and radionuclide concentrations in the water supply
- Radionuclide removal efficiency for the system, and
- The amount of sludge produced *per* unit volume of water processed.

Water treatment sludges are placed in lagoons and may include lime sludge, back flush water, spent ion-exchange media, and sand filter elements. Disposal in lagoons results in the accumulation of radium in bottom sediments that may have to be dredged and disposed of properly. Sludge is also disposed of in sanitary landfills, discharged to sewers, injected in deep wells, or spread on agricultural soils, while the decanted water is recycled.

Radon fluxes from disposed sludges are assumed to be near those of typical soils. Radiation exposure rates from sludges are expected to be near those of ambient background levels. Exposure rates from spent resins and charcoal beds, however, would be much higher.

Exposure levels as high as several  $\text{mR hr}^{-1}$  have been observed on charcoal and resin beds. An average of  $\sim 0.748 \mu\text{Sv hr}^{-1}$  ( $86 \mu\text{R hr}^{-1}$ ) was adopted for the referenced report.

## Metal Mining and Processing Waste

The mining and processing of ores for the production of metals generates large quantities of residual bulk solid and liquid wastes. Because the minerals of value make up only a small fraction of the ore, most of this bulk material has no direct use. It is estimated that the mining and processing of ores and minerals, other than uranium and phosphate, has resulted in the production of more than 40 billion MT (44 billion short tons) of mine waste and tailings from 1910 to 1981.

The metals extraction industry typically generates about 1.5 billion MT (1.65 billion short tons) of waste *per year*, including about 1.0 billion MT (1.1 billion short tons) of waste rock and overburden, 0.40 billion MT (0.44 billion short tons) of ore tailings, and less than 0.10 billion MT (0.11 billion short tons) of smelter slag. Depending on the original ores and processing methods, some of these wastes contain elevated concentrations of TENORM (Table 15).

It is generally believed by geologists that the level of NORM found in ores depends more on the geologic formation or region rather than on the particular type of mineral being mined. These ores often contain many different minerals, and the radionuclide content of one type of ore or mining operation or its wastes will not be representative of other mines or waste types. For some ores, the refining process may yield a waste process that may contain higher radionuclide concentrations when compared to the original ore. It has been reported that some of the more uncommon metals have highly radioactive waste products. Also, some processes associated with metal extraction appear to concentrate certain radionuclides and enhance their mobility.

Most of the metal mining waste is stored on-site or near the point of generation, in tailings ponds or used to construct dams, dikes, and embankments. About two-thirds is mine waste, and one-third is tailings. Metal mining processing wastes have only been reused in a limited number of applications, typically for backfilling mined out areas and for construction and road building near the mines. Some mineral processing wastes have been used to make wallboard and concrete.

Some of the mining wastes are stored in stockpiles that are reprocessed several times to extract additional minerals. NRC staff published guidance on September 22, 1995 (NRC 1995); allowing for certain feedstocks containing uranium and thorium to be processed by licensed uranium mills. This will allow the wastes to be disposed of in the uranium mill tailings pile. There are several restrictions on the feedstock.

**Table 15. Metal and Mining Industries Known or Believed to Involve TENORM**

Bauxite	Lead	Thorium
Beryllium	Molybdenum	Tin
Columbium	Nickel	Uranium
Copper	Rare Earths (Lanthanides)	Titanium
Gold	Silver	Zinc
Iron	Tantalum	Zirconium

Source: EPA (1993)

### **Rare Earths**

The rare earth elements, sometimes called lanthanides, are a group of 15 chemical elements with atomic numbers 57 through 71. Yttrium, which has an atomic number of 39, is also included because it occurs with other rare earth elements and has similar chemical properties. The special properties of the rare earth elements are why they are used in catalysts, ceramics, refractory and metallurgical processes, magnets, *etc.* They are also used in low-temperature superconductor technology, which may increase their demand in the future. The United States is the world's leading producer of rare earth elements. Rare earth oxides include bastnasite, monazite, and xenotime.



Bastnasite (also spelled bastnaesite) can contain up to 75% rare earth oxides, including up to 0.1% ThO<sub>2</sub>. Monazite can contain about 60% rare earth oxides, including 4 to 10% ThO<sub>2</sub>. Uranium may also occur in monazite at 0.1 to 0.5% U<sub>3</sub>O<sub>8</sub>. Thorium can be removed from monazite ores by several methods, resulting in thorium residue wastes. Xenotime can contain elevated levels of thorium and uranium. The ThO<sub>2</sub> and U<sub>3</sub>O<sub>8</sub> components from the rare earth metals appear in the waste products. Although some of these wastes have been treated as low-level waste and disposed of properly, some of the TENORM-contaminated wastes remain at the processing sites.

The annual generation rate of waste is assumed to be 20,800 MT (22,900 short tons) *per* year containing 6% TENORM with relative activities of 144 Bq g<sup>-1</sup> (3,900 pCi g<sup>-1</sup>) for thorium and 666 Bq g<sup>-1</sup> (18,000 pCi g<sup>-1</sup>) uranium. These values are considerably higher than the NRC's 0.05% for source material.

The radon flux rate from rare earth oxide waste piles depends on many factors, such as the radium concentration in the wastes, moisture content, porosity, and depth of the pile. The radon emanation coefficient for these wastes is estimated at 0.3.

Radiation exposure rates associated with these wastes can range from near background to several μSv hr<sup>-1</sup> (several hundred μR hr<sup>-1</sup>) for monazite wastes. Depending on the source, radiation levels may differ because many of the decay products may no longer be in secular equilibrium with uranium and thorium. A total external radiation exposure rate from thorium and uranium can be up to ~122 μSv hr<sup>-1</sup> (14 mR hr<sup>-1</sup>).

### Other Metals

Zirconium, hafnium, titanium, and tin generate approximately 470,000 MT (518,000 short tons) of waste a year with an average <sup>226</sup>Ra concentration of 1.59 Bq g<sup>-1</sup> (43 pCi g<sup>-1</sup>). Much of the ore from which titanium is obtained originates in sands that also contain monazite. Ores can contain concentrations of uranium and thorium in the range of 0.18 to 0.74 Bq g<sup>-1</sup> (5 to 20 pCi

$\text{g}^{-1}$ ). Total radium in sludge from titanium process streams had concentrations as high as  $2.85 \text{ Bq g}^{-1}$  ( $77 \text{ pCi g}^{-1}$ ). Some  $\text{ZrO}_2$  concentrates from South Africa are used in a process that chlorinates the sands and converts the zirconium to tetrachloride.

Measurements indicate that  $^{226}\text{Ra}$  concentrations in this ore are about  $7.4 \text{ Bq g}^{-1}$  ( $200 \text{ pCi g}^{-1}$ ). Direct chlorination of zircon puts the radium into the highly soluble radium chloride chemical form, which can yield high leachate concentrations in liquid waste streams. Values of  $1665 \text{ Bq L}^{-1}$  ( $45,000 \text{ pCi L}^{-1}$ ) of  $^{226}\text{Ra}$  were detected in water samples at one plant. The high solubility and mobility of radium chloride could pose a potential threat to the environment.

Amang is a general term for the by-products obtained when tin tailings are processed into concentrated ores. It includes minerals such as monazite, zircon, ilmenite, rutile, and garnet. Radium-226 and  $^{232}\text{Th}$  activities in amang have been reported to range from  $15.91$  to  $17.76 \text{ Bq g}^{-1}$  ( $430$  to  $480 \text{ pCi g}^{-1}$ ) and  $42.9$  to  $326.7 \text{ Bq g}^{-1}$  ( $1,160$  to  $8,830 \text{ pCi g}^{-1}$ ) respectively. Tailings from these ores may have a significant potential to cause elevated radiation exposures.

Measurements made at a tin smelter showed  $^{238}\text{U}$  concentrations up to  $1.59 \text{ Bq g}^{-1}$  ( $43 \text{ pCi g}^{-1}$ ) and  $^{232}\text{Th}$  concentrations up to  $0.7 \text{ Bq g}^{-1}$  ( $19 \text{ pCi g}^{-1}$ ). Gamma survey measurements at a tin smelter showed radiation levels in slag storage areas ranging from  $\sim 0.087$  to  $4.35 \mu\text{Sv hr}^{-1}$  ( $10 \mu\text{R hr}^{-1}$  to  $500 \mu\text{R hr}^{-1}$ ), with average levels less than  $\sim 0.522 \mu\text{Sv hr}^{-1}$  ( $60 \mu\text{R hr}^{-1}$ ). The large industries, including copper and iron, generate over 1.0 billion MT (1.1 billion short tons) of waste *per year*, with an average  $^{226}\text{Ra}$  concentration of  $0.18 \text{ Bq g}^{-1}$  ( $5 \text{ pCi g}^{-1}$ ).

## Geothermal Energy Production Waste

Geothermal energy in the United States is utilized only in a few places, mostly in California. Solid wastes originating from the treatment of spent brines contain TENORM. Hot, saline fluids from geothermal reservoirs may have a dissolved solids content approaching 30% by weight. The average  $^{226}\text{Ra}$  concentration in this waste is estimated at  $4.88 \text{ Bq g}^{-1}$  ( $132 \text{ pCi g}^{-1}$ ), with waste generation estimated at 54,000 MT (59,500 short tons).

## REGULATION, CONTROL, AND MANAGEMENT OF TENORM

### Federal Regulation of TENORM

EPA and other Federal and State agencies are responsible for regulating public exposures to NORM that are not licensed by NRC. State authority is derived from the Constitution, by which the States have primary responsibility for the health and safety of the public. EPA, State, and NRC programs do not treat the radiological risks from NORM consistently. NRC licensees generally are required to meet more restrictive conditions than are possessors and users of other NORM. There are no significant differences in the radiological risks of these materials, although radon and some discrete radium sources have a higher radiological hazard than uranium and thorium (NRC, 1996).

The definition of source material found in the Atomic Energy Act (AEA 1954) is based on the early safeguards concerns for material that could be used to ultimately make reactor fuel or nuclear weapons. When the definition was written, Congress considered that source materials needed to be placed under regulatory control on the basis of promoting common defense and national security. The health and safety impacts from NORM other than source material were considered to be manageable, to be relatively insignificant, and to have no basis for regulation from the standpoint on the common defense and national security (NRC, 1996).

The hazards posed by mill tailings (by-product materials) were incompletely recognized in the uranium industry's early years, and, while the AEA of 1954 instituted licensing of mill operators, tailings remained free of controls (EPA 1980). Byproduct material under the Act limited control to tailings "produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content" (AEA, 1954). Therefore, other tailings (vanadium, radium, *etc.*) are not regulated by the AEA, and are considered TENORM.

In 1965, it was discovered by the Public Health Service (PHS) and the Colorado Department of Health that uranium mill tailings were being hauled from the mill site and used

for construction purposes in around habitable structures (CDH, 1989). Regulations were promulgated to effect cleanup for Grand Junction based on PHS recommendations, known as the Grand Junction Remedial Action Criteria, found at 10 CFR 712 (AEC, 1972). These regulations were designed to mitigate radon in structures from uranium mill tailings. In 1978, the Uranium Mill Tailings Radiation Control Act (UMTRCA) was passed to address the mill sites themselves, as well as disposal of the tailings. The regulations supporting UMTRCA are found at 40 CFR 192 (EPA, 1980) (Table 16). These regulations are the basis for the current regulations for NORM the States have adopted, along with surface contamination release limits found in NUREG 1.86 (NRC, 1974).

**Table 16. 40 CFR 192 Standards**

Soil, $^{226}\text{Ra}$ averaged over 100 $\text{m}^2$ , shall not exceed background by more than:	<p>5 <math>\text{pCi g}^{-1} \text{ }^{226}\text{Ra}</math> averaged over the first 15 cm of soil below the surface</p> <p>15 <math>\text{pCi g}^{-1} \text{ }^{226}\text{Ra}</math> averaged over 15 cm thick layers of soil more than 15 cm below the surface</p>
Habitable buildings:	<p>Annual average radon decay product concentration (including background) not to exceed 0.02 WL. In any case, not to exceed 0.03 WL</p> <p>Level of gamma radiation shall not exceed the background level by more than 20 microrentgens <i>per</i> hour.</p>

There are a number of issues to be considered when adopting the 40 CFR 192 values to TENORM:

- The limits were promulgated when radiation protection guidance policies in place at that time limited exposures to the public to  $\sim 5 \text{ mSv yr}^{-1}$  ( $500 \text{ mrem yr}^{-1}$ ) whole body with limiting factors to critical organs. The proposed RPG is for an upper limit of  $\sim 1.0 \text{ mSv yr}^{-1}$  ( $100 \text{ mrem yr}^{-1}$ ) from all sources (60 CFR 49296).
- The risks from low levels of radiation are assumed to be proportional to dose, that is, they are based on the linear no-threshold model. There is considerable debate over the validity of this theory (Patterson, 1997).
- The limits in 40 CFR 192 were calculated using radon emanation values for sandy material. Many TENORM wastes have very low radon emanation fractions (slag).
- The indoor gamma exposure rate criteria of  $\sim 0.174 \text{ mSv hr}^{-1}$  ( $20 \text{ mR hr}^{-1}$ ) above background was designed to allow some limited flexibility in the methods chosen to reduce indoor radon decay product concentrations, not to meet a certain dose limit. In fact, based on 75% occupancy, the standard would allow gamma radiation doses from the tailings of about  $\sim 1.13 \text{ mSv yr}^{-1}$  ( $130 \text{ mrad yr}^{-1}$ ) (EPA, 1980).

***Currently there are no federal regulations specifically controlling TENORM.***

EPA was going to consider TENORM in proposing 40 CFR 196, but that rule was withdrawn at the request of DOE. It is unlikely that TENORM would be in a final rule due to pressure from industry.

*Def - Not AEA*

## States Regulation of TENORM

Many states consider TENORM to be regulated by their general rules on radiation. Other States believe that TENORM should have specific regulations. The Conference of Radiation Control Program Directors (CRCPD) has developed templates for States to use in drafting regulations for control and disposal of TENORM. The previous drafts were based on the 40 CFR 192 radium in soil values with exemptions, methods for licensing, protection of workers and general population, and disposal. The draft regulations have gone through many iterations. Eight states currently have regulations pertaining to TENORM, most of them based on the CRCPD template. The States are listed in **Table 17**. CRCPD has established a blue ribbon panel to work more efficiently and effectively to finalize the Part N suggested state regulations for the control of TENORM. The panel released a new draft of the proposed State regulations in February 1997, the comment period ends June 30, 1997 (HPS 1997). A review of the new draft follows.

**Table 17. States with TENORM regulations**

New Mexico	South Carolina	Mississippi
Arkansas	Louisiana	Texas
Ohio	Georgia	Oregon

Other states currently considering TENORM regulations are listed in **Table 18**.

**Table 18. States considering TENORM regulations**

Alabama	Alaska		Connecticut	Florida
Illinois	Michigan	New Jersey	Oklahoma	Colorado?

## CRCPD Suggested State Regulations for Control of Radiation (Part N)

Some features of the current draft are:

- A new definition of what TENORM is: “naturally occurring materials not regulated under the AEA whose radionuclide concentrations have been increased by or as a result of human practices. TENORM does not include the natural radioactivity of rocks or soils, or background radiation, but instead refers to materials whose radioactivity is technologically enhanced by controllable practices (or by past human practices)”.
- The limits in the standard are dose-based. The implementing State is to determine what fraction of 100 mrem yr<sup>-1</sup> TEDE (excluding natural background) to the reasonably maximally exposed individual is allowed from TENORM.
- Exemption limit of 5 pCi g<sup>-1</sup> <sup>226</sup>Ra or <sup>228</sup>Ra,
- Surface contamination guidelines follows NUREG 1.86 (NRC 1974),
- Excludes indoor radon from TEDE calculations,
- States are given a flexibility for implementing Part N consistent with their respective, unique circumstances,
- Safety criteria for products containing TENORM,
- Quality control, labeling and reports of transfer of TENORM,
- Implementation Guidance will be developed that will address issues such as determination of background, survey methods, *etc.*

✓ Not  
Dose  
Based

**HPS/ANSI Standard for NORM - Guide for Control and Release of NORM**

In addition to the CRCPD efforts, the HPS has a working group that is developing an ANSI standard for control and release of NORM (HPS, 1997a). The working group is comprised of representatives of industry and government. The standard is still in draft form, consensus has not been reached on all issues, however, some basic themes of the standard can be discussed (Dehmel, 1997):

- Primary exposure limit of 1 mSv (100 mrem)  $\text{yr}^{-1}$ . TEDE, above background to average member of critical group exposed under realistic conditions, does not include radon,
- Limit to be calculated over 1,000 years,
- Allows for institutional or engineered controls,
- Provisional limit for infrequent exposures to RME of 5 mSv (500 mrem)  $\text{yr}^{-1}$  during remediation of facilities contaminated by past practices,
- Surface guidelines adopted from draft ANSI N13.12, July 1996 draft,
- Outdoor radon limited to 20  $\text{pCi sec}^{-1} \text{ m}^{-2}$ , averaged over the entire area of the disposal unit, waste or material pile, or impoundment,
- Indoor radon limited to 4  $\text{pCi L}^{-1}$  in areas that are occupied or occupiable,
- Dose limits for products or materials containing NORM.



## **GUIDANCE DOCUMENTS FOR TENORM**

In addition to the CRCPD template for State regulations, some guidelines for the control, disposal, and release of TENORM are:

- *Guidelines for the Handling of NORM in Western Canada* (WCNC, 1995),
- Implementation manual for Management of NORM in Louisiana (LDEQ, 1990).
- Texas also has published regulatory guides on conducting close-out surveys of open land areas and requesting release for unrestrictive use (BRC, 1990).
- *Management of NORM in Oil and Gas Production*. (API, 1993) by the American Petroleum Institute.
- *Radiation Protection in the Mineral Extraction Industry*. NCRP Report No. 118.

## **RECYCLING AND DISPOSAL OF TENORM**

Reuse of contaminated scrap metal is an industry unto itself and is the topic of much discussion. Scrap dealers and smelting facilities have detected the presence of radioactivity, including TENORM, in numerous shipments of scrap metals by the use of radiation detectors at their facilities. More sensitive and rugged detector systems are currently in development for metal recycling facilities and similar facilities. These should help to protect these industries from accidental recycling radioactive materials such as TENORM and anthropogenic sources.

Envirocare of Utah owns a licensed facility for commercial TENORM disposal located in Clive, Utah. The licensing of this facility follows criteria similar to those pertaining to uranium mill tailings disposal.

The U.S. Ecology low-level waste facility at Hanford, Washington will accept some TENORM wastes, but with restrictions. Extra packaging, waste form, and design requirements may result in lower radon releases and waste leach rates. This option would be limited by cost and volume restrictions.

Newpark Environmental TENORM Processing Facility of Port Arthur, Texas accepts TENORM wastes for processing for injection into deep wells.

Campbell Wells Corporation of Lafayette, Louisiana accepts TENORM and NOW for treatment and disposal.

Efforts have been made to convince NRC to allow disposal of TENORM wastes in 11e.(2) disposal cells. NRC staff published a notice in the Federal Register on September 22, 1995, stating that "Radioactive material not regulated under the AEA shall not be authorized for disposal in an 11e.(2) byproduct material impoundment" (NRC 1995).

## EXAMPLES OF TENORM EXPOSURES

There have been a number of cases where the improper disposal of TENORM wastes has resulted in increased levels of direct radiation exposure to individuals. A few examples include:

- In Montclair, New Jersey, radium-contaminated soil caused elevated gamma exposure rate levels. This project is now a CERCLA site; cleanup is under way (EPA 1990).
- In Polk County, Florida surface soils have been removed during grading at construction site to expose and redistribute low-grade phosphate ores. Houses and condomenia have been built directly on these deposits. No studies of  $^{222}\text{Rn}$  fluxes in the structures built at these sites are known to have been carried out. Typical radon fluxes in re-worked surface deposits near this site are quite elevated; sometimes by an order of magnitude or more than local backgrounds.

- Elemental phosphate slag used to construct roads in Pocatello, Idaho, has resulted in a doubling of the radiation levels in some areas. Phosphate ore tailings have also been used as aggregates on dirt roads in some counties in Florida. No studies have directly addressed the increased exposure to the public due to this activity.
- In Mississippi, recycled pipes that are contaminated with radium scales have been used to construct playground equipment and are used in welding classes. Both activities have resulted in unnecessary exposures.
- A phosphate fertilizer facility in Louisiana discharged thousands of tons a day of phosphogypsum and phosphate processing effluents directly into the Mississippi River. This practice ceased in the mid-1980's. Similar types of discharges in Spain have been found to significantly increase  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$ , and U isotopes in rivers downstream of phosphate processing facilities.
- Vanadium and radium tailings have been used in construction materials and have contaminated soil and groundwater.
- In the past, pipe scale residue was left on the ground at pipe cleaning yards or washed into ponds or drainage basins. Surveys showed that some locations exhibited external radiation levels above  $2 \text{ mR hr}^{-1}$  and  $^{226}\text{Ra}$  concentrations above  $1,000 \text{ pCi g}^{-1}$ .
- Oil field sludges often were dumped into waste pits. Both burn and brine waste pits have been used for disposal of sludges and production water residues. This past practice may lead to ground and surface water contamination. In addition, direct radiation exposures may have occurred to individuals working or living near the disposal pits.

- Relatively elevated concentrations of  $^{226}\text{Ra}$  were used for decades to produce luminous dials on instruments in aircraft, military equipment, on watches, *etc.*

In addition to these specific examples, numerous incidents occur each year in metal recycling. Scrap metals containing elevated TENORM as well as radiation sources are inadvertently combusted and smelted. These examples represent a very small fraction of events that result in elevated exposures, either through ignorance or neglect, to TENORM that the public is subjected to on a regular basis.

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## **APPENDIX A**

### **TERMS AND DEFINITIONS**

#### **NORM and TENORM Producers, Users, and Proposed Regulations**

**HPS PEP Course 1.A**

**24 January 1999**



## APPENDIX A

### TERMS AND DEFINITIONS

**Atomic Energy Act Definitions ( 42 USC 1954):** Source material: Sec.11(z) "... means (1) uranium, thorium, or any other material which is determined by the Commission pursuant to the provisions of section 61 [42 U.S.C. 2091] to be source material; or (2) ores containing one or more of the foregoing materials, in such concentrations as the Commission may by regulation determine from time to time" (0.05% by weight).

Special nuclear material: Sec.11(aa) "... means (1) plutonium, uranium enriched in the isotope 233 or in the isotope 235, and any other material which the Commission, pursuant to the provisions of section 51 [42 U.S.C. 2071], determines to be special nuclear material, but does not include source material; or (2) any material artificially enriched by any of the foregoing, but does not include source material."

By-product material: Sec.11(e) "... means (1) any radioactive material (except special nuclear material) yielded in or made radioactive by exposure to the radiation incident to the process of producing or utilizing special nuclear material, and (2) the tailings or wastes produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content."

**NARM:** Naturally Occurring and Accelerator Produced Material. Any radioactive material that can be considered naturally occurring and is not source, special nuclear, or by-product material or that is produced in a charged particle accelerator (DOE 1988).

**NORM:** Naturally Occurring Radioactive Material, NORM is considered a subset of NARM. NORM is basically defined by exclusion. This has caused regulatory problems because NORM is not specifically regulated by the AEA. Definitions vary between agencies and all have exemptions.

**HPS (1997a):** "...means any radionuclides or radioactivity disturbed by man-made activities or technologically-enhanced state, which may result in a relative increase in radiation exposures and risks to the public above background radiation levels.

**Technologically enhanced:** "...means that the physical and chemical properties have been altered and radionuclide concentrations have been increased by human practices, such that there exists a potential for:

- 1) Exposures to individuals or populations
- 2) Environmental redistribution and contamination
- 3) Increased environmental mobility
- 4) Incorporation of radioactivity in products and construction materials
- 5) Recycling or re-use of contaminated material or equipment
- 6) Improper disposal or use of disposal methods that could result in unnecessary exposures to individuals and populations or environmental contamination

**Technologically Enhanced Naturally Occurring Radioactive Material: CRCPD (1997):**

"...means naturally occurring materials not regulated under the AEA whose radionuclide concentrations have been increased by or as a result of human practices. TENORM does not include the natural radioactivity of rocks or soils, or background radiation, but instead refers to materials whose radioactivity is

technologically enhanced by controllable practices (or by past human practices).

**Technologically Enhanced Naturally Occurring Radioactive Material: (cont'd.)**

Gesell and Prichard (1975) define technological enhancement as: "...exposures to truly natural sources of radiation (*i.e.*, naturally occurring isotopes and cosmic radiation) which would not occur without (or would be increased by) some technological activity not expressly designed to produce radiation."

There are two methods to consider under their definition:

- 1) Bringing the receptor to the source; *e.g.*, inadvertent or deliberate proximity to a radiation source, air or space travel
- 2) Bringing the source to the receptor:
  - Industrial processes
  - Consumer products
  - Indoor radon

Risk - Modeling - Bounding - Relatively Easy  
- Realities - Very Hard  
- Outcome Curve.

5th - One hour 500 m/yr - Practical 2 m/yr 100 m/yr  
100 m/yr -  
- ICRP 30 m/yr per year constraint  
- EPA 15 m/yr Limit

1 pCi/gm U - Ra  $\sim$  20 mrem/yr Breckenridge Model

5 pCi/gm Ra  $\sim$  75 mrem/yr. Area

Map  $\rightarrow$  Configure over life.

Can not regulate TENORM at the level leading to  
regulate AEA material

Iceland - High Risk - Geothermal Water.

Commission

- Exemption 40.13(a) 500 ppm
- Amer Mini A - Mill Tailings - Friction
- Proposed Rule - Decom - U/T for - 5 pCi/gm Ra Benchmark  
     $\rightarrow$  Practical 9th.
- Interface TENORM - AEA 5th.

Yes - Yellowstone - K<sub>2</sub>Cl  
- Reduced System.